Atmospheric Pollution Research 2 (2011) 80-88

Atmospheric Pollution Research

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Analysis of the relationship between urban background air pollution concentrations and the personal exposure of office workers in Dublin, Ireland, using baseline separation techniques

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ABSTRACT

Real–time concentrations of PM_{10} were monitored over a 24 hour period for a number of different subjects as part of an investigation to examine the influence of daily activities and locations on the personal exposure of city centre office workers to air pollution. The resulting data comprised time series plots consisting of a series of peaks and troughs as a result of exposure to the differing sources of particulate matter subjects were encountering as well as the underlying background concentration. In order to separate the background concentration component from the data a number of baseflow separation techniques were employed, commonly used in stream–flow hydrology. Filter separation and frequency analysis techniques were examined comparing their predictions of background concentration with urban background concentration measurements for reference. The results of this investigation highlight a number of different approaches to separating background concentration from real–time personal exposure data. These methods will enable further investigation of purely activity and location based personal exposures as well as improvements in the numerical modelling of air pollution exposure in future. The results of this investigation also demonstrate a novel synergy in methods of analysis between the fields of air pollution and hydrology.

Keywords:
PM_{10}
Personal exposure
Background concentration
Baseline separation
Air pollution

Article History:
Received: 05 August 2010
Revised: 23 September 2010
Accepted: 19 October 2010

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doi: 10.5094/APR.2011.010

1. Introduction

In recent years, research in the field of air pollution and human health has begun to focus its attention more on the investigation of personal exposure. Measurements of personal exposure to particulate air pollution have been shown to have more direct links with adverse impacts on human health compared to background concentrations (Seaton et al., 1995; Schwartz et al., 1996; Pope, 2000; Dockery, 2001). Therefore the previous traditional and current regulatory monitoring of background air pollution concentrations has seen a shift in terms of health assessment to personal exposure measurements.

The average daily personal exposure experienced by a typical urban office worker is a multifaceted conglomeration of the effects of the numerous sources of air pollution the typical individual experiences on a daily basis. In an attempt to better understand the daily personal exposure of office workers, an investigation is underway, the PALM project (Personal–exposure, Activity and Location Model), in Dublin Ireland whereby real–time personal exposure to particulate matter is being monitored for various subjects while also monitoring their activities and locations (McCreddin et al., 2009). These data then facilitate the derivation of different components of personal exposure according to the activity and/or location of the subject in question. The analysis of these components of personal exposure is expected to produce the capability for better predictions of personal exposure to air pollution in future and a method of modelling personal exposure based on activity and location.

Of the numerous components of exposure being investigated in the PALM project (such as transport emissions, indoor air emissions, environmental tobacco smoke, point sources, etc.) the background concentration at any given location is an ever present contribution to the overall personal exposure of an individual regardless of the activities they are performing. Therefore it was deemed necessary to investigate methods of extracting the background exposure component from the real–time personal exposure measurements. The extraction of this data would enable: the assessment of the contribution of background air pollution to overall personal exposure, comparisons of the contribution of background and non–background exposure components, and subsequently better prediction of personal exposure overall.

Background concentration has been defined as the concentration of air pollution in the atmosphere at any one location which is not directly affected by local emission sources (Tchepel et al., 2010). Background concentration is however not a fixed value and varies in that it may be influenced by regional air quality and indirectly by local sources. Emissions of pollutants from neighbouring cities may travel long distances to influence the background concentration of another city on a regional scale (Beelen et al., 2009). Background concentration may also vary from hour to hour where it is indirectly influenced by local emissions i.e. background concentration is likely to increase in response to peak traffic emissions or decrease at night in response to minimal traffic emissions (Moreno et al., 2009). Background concentration has also been shown to vary on a spatial as well as temporal scale. The background concentration of air quality is likely to be considerably different on a large spatial scale between urban, sub–urban and rural environments due to lower rates of local emission in less densely populated areas (Beelen et al., 2009). Furthermore the background concentration in the different microenvironments people regularly pass between (e.g. office, home, outdoor urban, outdoor sub–urban, etc) is also likely to vary. Previous investigations have regularly found concentrations of air pollution indoors which is lower than the outdoor background concentration (Colome et al., 1992).

Previous investigations have been carried out which investigated the relationships between personal exposure and background concentrations using various techniques (Ballesta et al., 2008). However, the problem presented in extracting the contribution of background concentration from a 24–hour time series of personal exposure data was noted to be similar in nature to that of baseflow separation in the field of stream–flow hydrology. Here the contribution of groundwater is required to be separated from a time series of overall stream discharge to assess the contribution of surface run–off to a storm flow (Ekhardt, 2008). In the field of hydrology the flow of water in a stream is often assumed to comprise a component of flow associated with surface run–off and a component of flow associated with baseflow (flow from groundwater) (Aksoy et al., 2009). Surface run–off can be described as ”quick–response flow” which results in rapidly occurring spikes in the time series flow record while the baseflow produces a more steady response due to the slow nature of flow through aquifers and is thus ”slow–response flow”. Like background concentration, baseflow varies both temporally and spatially and is indirectly influenced by “local emissions” of precipitation. Baseflow is likely to increase in response to a local rainfall event or decrease in response to dry periods.

Some similarities therefore exist in the relationships between surface run–off/baseflow and personal/background exposure concentrations. Personal exposure is susceptible to the effects of various air pollution sources and as a result, presents a series of rapid response spikes in its time series history, personal exposure could be described as ”quick–response exposure”. Background concentration however is slow to respond to instantaneous increases in local air pollution concentration and instead provides a steady response to the overall air quality of the locality which could be described as “slow–response exposure”. Clearly fundamental differences exist in the underlying mechanics of the two relationships, however the two are certainly analogous to a certain degree. Therefore it was assumed that an adaptation of baseflow separation techniques to air pollution time series data could provide useful predictions of background air pollution. Numerous such methodologies exist in the field of hydrology and these have been investigated for their performance in the prediction of baseflow by numerous investigators (Bougthon, 1988; Chapman and Maxwell, 1996; Brodie and Hostetler, 2005; Ekhardt, 2005; Ekhardt, 2008; Aksoy et al., 2009). This paper presents an investigation of these methodologies to enable this extraction to be carried out using adaptations of baseflow separation techniques commonly used in the study of stream–flow hydrology.

2. Methodology

2.1. Personal exposure monitoring

Real time personal exposure sampling of PM_{10} was carried out using a Metone, Aerocet 531 aerosol profiler (MetOne Inc, 2003). The Aerocet 531 is a real–time photometric sampler, an automatic instrument that estimates PM in a range of 1, 2, 5, 7 and 10 µm in aerodynamic diameters. The instrument uses a right angle scattering method at 0.780 µm. The source light travels at a right angle to the collection system and detector, and the instrument uses the information from the scattered particles to calculate a mass per unit volume. A mean particle diameter is calculated for each of the five different sizes (Kumar et al., 2007). This mean particle diameter is used to calculate a volume (cubic meters), which is then multiplied by the number of particles and then a generic density (µg/m^3). The resulting mass is divided by the volume of air sampled for a mass per unit volume measurement (µg/m^3). The sampler was used to record concentrations of PM_{10} at 2 minute intervals over a 24 hour period. The Aerocet–531 was chosen because it is a portable handheld device, weighing approximately 0.88 kg, which made it extremely convenient for use in a personal exposure study of this nature where numerous volunteers were required to carry the device on their person for 24 hours. Ten samples were recorded between February and July 2009 by 6 separate volunteers. Each of the volunteers lived in the greater Dublin area and worked in an office environment in the city centre. Figure 1, shows a typical 24–hour time series profile recorded during this investigation.

During sampling, the location of each subject was also monitored using a GPS (Garmin GPSMAP® 60CSx) tracking device which each volunteer kept on their person at all times. Figure 2 shows a plan view of the 24 hour location pattern of one of the volunteers during their sampling. The sampling volunteers were instructed to keep the sampling kit (GPS & Aerocet 531) on their person at all times during the 24–hour sampling period. A small satchel was employed during sampling to house the sampling equipment together and prevent interference from the subjects. Volunteers were also instructed to complete a simple time series diary of their activities during the day (e.g. 8 am – 9 am, commuting by car; 9 am – 10 am in office; etc).

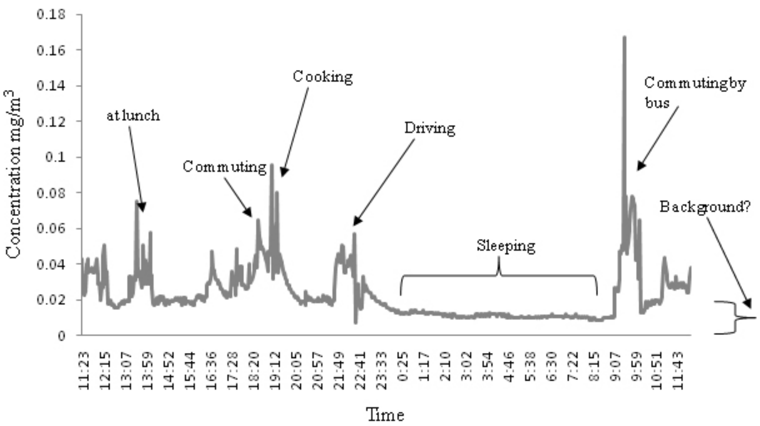

Figure 1. *Typical 24 hour time series profile of personal exposure to PM_{10}.*

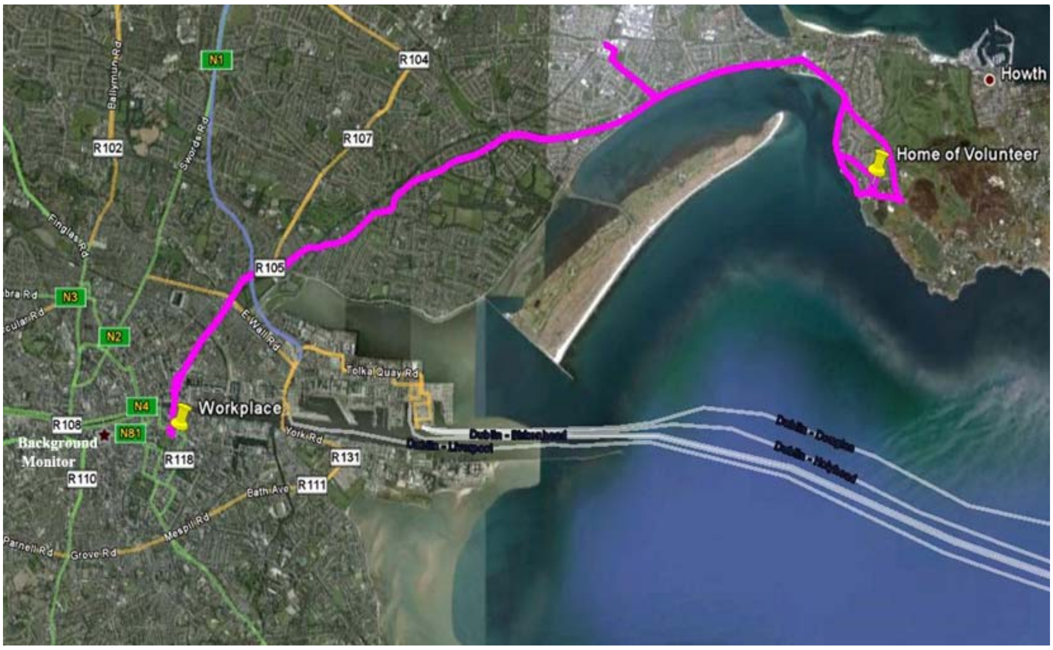

Figure 2. *Typical map of volunteer movements during sampling.*

2.2. Sampling protocol

Sampling was carried out according to the following protocol to ensure a high level of quality control and to ensure robustness in the data analysis:

- Volunteers fitting the definition of an office worker living in the greater Dublin area who commute to their workplace located in the city centre were recruited for sampling.
- Before sampling commenced the Aerocet–531 monitor was tested with a zero filter and flow meter to ensure proper functioning of the monitor.
- Each volunteer was given an Aerocet–531 monitor including a plug for charging (as the monitor had a battery life of approximately 8–hr), a handheld Garmin GPS device to record their movements, spare rechargeable batteries for the GPS, and finally, a log in which to record their activities while sampling.
- The monitor then recorded continuously the exposure of the volunteers for the duration of the sampling period.
- Volunteers were carefully instructed on how to fill the activity log giving as much detail as possible.
- Volunteers were also carefully instructed on the operation of the Aerocet monitor and GPS tracker. Volunteers were encouraged to refrain from intervening in the operation of both systems in general and are instructed on how to charge both devices and switch them on/off in case of accidental power–off.
- At the end of the sampling period the equipment was returned and the data from both the Aerocet and GPS was downloaded to a PC. The GPS data, along with the activity log, was then used to break the data from the Aerocet down into particulate exposures due to each different activity. This data along with various summary statistics and influencing variables (weather, traffic, subject descriptions, etc) were then fed into the overall dataset for the PALM project.

2.3. Quality control procedures

Prior to sampling, each personal exposure sampler was calibrated against a Haz–Dust EPAM–5000 particulate monitor which utilised the gravimetric technique employed in background concentration monitoring (see Section 2.4). It was important to compare the different methods of particulate sampling in order to establish the degree of correlation between them. These accuracy calibration experiments were carried out in both indoor and outdoor environments to assess any differences in the performance of the Aerocet–531 across a range of emissions sources, concentrations and atmospheric conditions. The indoor accuracy calibration was carried out in an office environment located Dublin City centre while the outdoor calibration was carried out on the busy roadside environment of Pearse Street, Dublin. During outdoor samples the monitoring equipment was located at head height on the footpath, 3 m from the roadside. During all accuracy calibration experiments the Aerocet–531 sampling inlet was located beside the inlet of the Haz–Dust EPAM–5000 particulate monitor and both instruments recorded samples in parallel for a period of 8 hours. Figure 3 shows the results of the PM_{10} calibration experiments, where it can be seen that good agreement between the two monitoring techniques was achieved.

The resulting calibration equation was then used to adjust the measurements taken by the Aerocet–531 to give an agreement between the two methods of over 79%. This level of agreement was deemed satisfactory.

The repeatability of the Aerocet–531 measurements was also assessed by employing several of the Aerocet–531 monitoring units to record a sample in the same environment simultaneously. Nine Aerocet–531 samplers were compared to one another over a number of 30–minute sampling periods located in both indoor and outdoor environments as described above. Analysis of this data was carried out to investigate if the deviations between mean concentrations recorded by the 9 sampling devices were statistically significant. A p–value of 0.488 was found inferring that this was not the case and therefore the 9 Aerocet samplers were deemed to provide sufficiently precise recordings.

Finally, the flow rate of the Aerocet–531 pump was required to be 2.83 L/min (±5%) and this was checked on a regular basis using a Dwyer flow meter. In addition, before any sampling periods commenced the Aerocet–531 sampler was tested for any leaks with a zero filter. The zero filter was attached to the Aerocet–531 inlet nozzle and removed 99.99% of all particles larger than 0.3 micron.

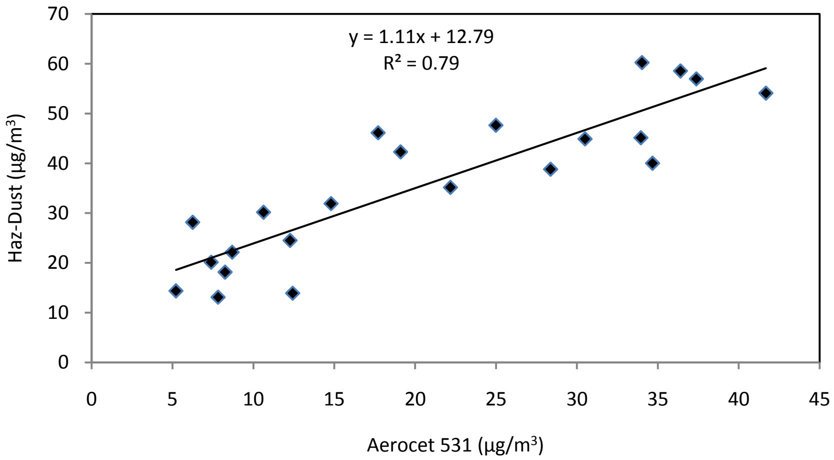

Figure 3. *Comparison of Aerocet-531 particle profiler and gravimetric method (Haz-Dust EPAM-5000) for indoor and outdoor city centre environments.*

2.4. Urban background concentration monitoring

Urban Background PM_{10} concentrations were also recorded by the local regulatory authority in Dublin city, reporting the 24–hour average PM_{10} concentration on each of the sampling days at a city centre location. This location at Winetavern Street was situated on the south side of the River Liffey approximately 500 m from the city centre (E: 315164.3, N: 234055.6) as shown in Figure 2. The monitoring station was located in accordance with schedule 8 of the Irish Air Quality Standards (S.I No. 271/2002) at the entrance to Dublin City Councils Civic, Offices, approximately 7 m from the roadside. The monitoring station was located to avoid measuring the concentration of very small micro–environments in its immediate vicinity and was representative of the air quality of at least the surrounding 200 m^2. The Winetavern Street station was also representative of other similar locations in Dublin city not in its immediate vicinity. The station therefore gave a measure of the background urban air quality typical of the outdoor urban environment around Dublin city centre which could be used to compare with predictions of background concentration from personal exposure samples for reference.

The samples were recorded by filtration and gravimetric analysis using an R&P Partisol (Rupprecht and Patachnick) through a PM_{10} inlet on 47 mm filters. These measurements were carried out in compliance with the reference method for the sampling and analysis of PM_{10} (CEN, 1999).

2.5. Descriptive statistics

The mean PM_{10} background air pollution concentration measured during this investigation was 23 µg/m^3, while the mean personal exposure to PM_{10} recorded was 32 µg/m^3. Comparing the average personal exposure recorded for each of the sampling days and the corresponding city centre background concentration revealed a directly proportional relationship between the two. As shown in Figure 4, personal exposure was found to increase with background concentration with a slope of 1.04. In terms of R^2, 35% of variation in the personal exposure concentrations was shown to be explained by the background air pollution levels. Considering the number of factors which influence air pollution concentrations, this represents as reasonably strong relationship between the two variables. However, it should be noted that only during the working day were the volunteers located within a reasonable distance to the background monitor (0–1 km). The mapping of volunteer movements (see Figure 2) and monitoring their daily activities showed that after working hours each subject tended to return to their respective suburban location until the following day. For roughly 16 hours of the 24 hour sampling period the volunteers could be located up to 14 km away from the back-ground monitoring station, thus reducing its influence on the overall 24 hour average personal exposures.

Comparing the average personal exposure of each subject over a typical 8–hour working day period (09:00–17:00) to the city centre background monitor revealed a significantly stronger relationship. A similar slope of 0.94 was found but the R^2 increased to 50%. It was therefore decided to use the baseflow separation techniques to separate the background concentrations from the personal exposure during the 8–hour working day period only as the PM_{10} background measurements were less appropriate for use with the entire 24–hour personal exposure time series. The predicted 8–hour background concentration obtained from the various separation techniques were then compared for accuracy and precision against the measured city centre background concentrations.

2.6. Baseflow separation

Baseflow separation uses the times series record of a stream–flow to derive the baseflow signature of a particular catchment (Brodie and Hostetler, 2005; Ekhardt, 2008). In the current investigation the time series record of personal exposure to PM_{10} was used to derive the background concentration "signature" of a particular sample.

Numerous methods of baseflow separation exist; these include graphical techniques, filtering methods, frequency analysis and recession analysis. Graphical techniques and recession analysis do not lend themselves to adaptation for use in air pollution studies due to differences in the nature of air pollution from stream–flow and due to the requirement for mathematical solutions to the problem to enable analysis of large sets of data in future. Frequency analysis and filtering methods were deemed suitable and the implementation of these is discussed further in the following subsections. Baseline separation was carried out in the present study using Microsoft Excel software.

2.7. Filter separation techniques

Filter separation techniques can be used on time series data to separate the background concentration components through data processing or filtering procedures. Four examples of these techniques were investigated for their effectiveness with air pollution data, namely the Boughton method, smoothed minima, sliding interval method and recursive digital filters. For each method, the predicted background concentrations for the 10 PM_{10} personal exposure samples analysed were compared with the measured urban background PM_{10} concentrations to determine the extent of the relationship between the two for reference.

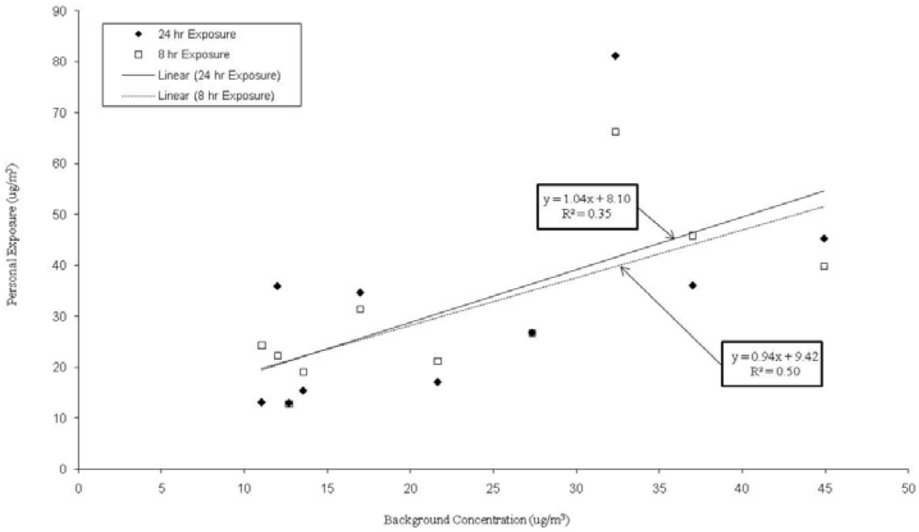

Figure 4. *City centre background PM_{10} concentration vs. mean PM_{10} personal exposure.*

Boughton method. The Boughton method was developed in hydrology to predict baseflow, whereby the baseflow was increased at each time step by either a constant rate or by a fraction of the run–off (Boughton, 1988). The Boughton method was modified for the purposes of the current investigation to predict background concentrations from personal exposure time series data. Background concentration data were predicted from each time series sample using a logic function. The function operated by increasing/decreasing the background concentration at each time step according to a recession constant times the previous personal exposure. The decision on whether to increase or decrease the background concentration at any given time step was based on whether the overall personal exposure was increasing or decreasing at that point. Also separate recession constants were used for increasing or decreasing the background prediction. Therefore the background concentration increased and decreased in line with the personal exposure but its rates of increase and decrease were separate. This separation technique would therefore follow the theory that background concentration is affected by changes local air quality but its response to these changes should be considerably lower in magnitude than the response of personal exposure. Furthermore the predicted background concentration at each time step was never allowed to be less than zero or greater than the measured personal exposure at that time. The magnitude of the recession constants *a* and *b* were subsequently optimised by trial and error to obtain the best fit relationship between measured reference and predicted background concentrations.

Smoothed minima. The smoothed minima technique uses the minimum stream–flow from the previous non–overlapping 5–day period as a measure of baseflow in hydrology (IOH, 1980). Here, this technique was used to determine the background concentration from the personal exposure data by assuming that the background concentration was equal to the minimum personal exposure value from a previous non–overlapping time period. The length of this time period was initially chosen as the previous non–overlapping 5–time series points which amounted to a 10–minute interval. The length of this time period was later optimised to obtain the best fit relationship between measured reference and predicted background concentrations. Again this method acts to dampen the response of personal exposure to changes in local air quality so that its response to these changes is "slow" making it more representative of the likely background air pollution response at any one time.

Sliding interval method. In hydrology the sliding interval method assigns a baseflow to a particular time step based on the lowest discharge value found within a fixed time period before and after that particular time step (Pettyjohn and Henning, 1979). In the current investigation this method was employed to obtain background concentration predictions in a similar manner to the smoothed minima method while using a sliding time period as opposed to a non–overlapping one. Again the results of this section of the analysis were optimised by trial and error by modifying the length of the sliding interval period i.e. the degree of damping of the personal exposure time series.

Recursive digital filters. Recursive digital filters (RDF) are routine tools in signal processing and they have also been used in hydrology to remove the high frequency quick flow signal to derive the low frequency baseflow signal (Nathan and McMahon, 1990). Numerous equations have been developed to smooth hydrographic data and three such equations have been used to investigate the effectiveness of RDFs in air pollution studies. The first RDF examined is shown in Equation (1) which is an adaption of the one–parameter filter developed by Eckhardt (2005):

$$E_{b(i)} = \frac{\left(1-E_{\max}\right)aE_{b(i-1)} + \left(1-a\right)E_{\max}E_i}{aE_{\max}} \tag{1}$$

where $E_{b(i)}$ is the background concentration at time *i*, E_{max} is the maximum exposure concentration in the time series, *a* is a recession constant, $E_{b(i-1)}$ is the background concentration at the previous time step, and E_i is the exposure concentration at time *i*. In this equation the recession constant *a* was modified by trial and error to obtain the best fit between measured reference and predicted background concentrations. The result of this equation produces a damped or lower frequency response to the personal exposure time series. However care must be taken to ensure that the predicted background concentration is always less than the measured personal exposure at each time step (i.e. $a < 1.0$) and greater than zero.

The second RDF examined, shown in Equation (2), is an adaption of another one–parameter filter called the one–parameter algorithm (Chapman and Maxwell, 1996). Nomenclature, similar to RDF 1, was used with the addition of *k*, another recession constant.

$$E_{b(i)} = \frac{k}{2-k}E_{b(i-1)} + \frac{1-k}{2-k}E_i \quad (2)$$

$$E_{b(i)} = \frac{k}{1+C}E_{b(i-1)} + \frac{C}{1+C}E_i \quad (3)$$

The last RDF examined, shown in Equation (3), is an adaptation of a two–parameter filter known as the Boughton two–parameter algorithm (Chapman and Maxwell, 1996). *k* and *C* are recession constants which were optimised by trial and error to obtain the best fit between measured reference and predicted data.

2.8. Frequency analysis

Frequency analysis presents a different approach to deriving background concentration data. In hydrology it has been used to determine the relationship between the magnitude and frequency of stream–flow discharge (Brodie and Hostetler, 2005). Here the relationship between personal exposure magnitude and frequency was determined by sorting each time series in order of decreasing concentration. Each time step was given a unique ranking number *m*, 1 for the maximum exposure concentration to *n* for the minimum concentration. The probability *P* of a concentration being equalled or exceeded was then obtained using Equation (4):

$$P = 100\frac{m}{n+1} \quad (4)$$

The calculation of *P* for a particular time series sample then enabled a concentration–probability plot to be drawn up. Figure 5 shows a typical concentration–probability plot obtained during the analysis. Examining these plots facilitated the formation of assumptions or hypotheses that the background concentration in any personal exposure time series could be obtained by assuming it is equal to the personal exposure concentration whose probability of occurring was 50 or 20% of the time.

The personal exposure for each sample at *P* = 50%, 40%, 20% and 10% were determined from the frequency analysis and these were subsequently compared with the measured reference background concentrations. The results of this comparison revealed the validity of assuming the P_x exposure concentration of a particular sample was equal to the mean background concentration.

3. Results

3.1. Filter separation techniques

Boughton method. The values of the recession constants *a* and *b* were altered to give the best fit between the predicted and measured background concentrations, using values of *a*=0.1 and *b*=0.45 produced the best results. Plotting the measured and predicted background concentrations (not shown) revealed an approximately linear relationship between the two where: *Boughton separated background = 0.99x(measured background) + 5.20.*

The Boughton separated background prediction was found to give a very good estimate of the measured concentration with a slope of 0.99 and the relationship between the two was found to account for a significant amount of variation in the data with an R^2 of 60%. Figure 6 shows a typical output of Boughton separated background concentration from a personal exposure time series plot. Due to the high values of recession constants used the separated background concentration produces a series of rapidly increasing/decreasing spikes in response to events in the personal exposure time series.

Smoothed minima. Using the smoothed minima filter separation technique, the personal exposure concentrations were "smoothed" over a specified non–overlapping time period. This time period was initially chosen at ten minutes, however as shown in Figure S1 in the Supporting Material (SM), this resulted in good accuracy between the predicted background concentrations and the measured values but poor precision. Increasing the interval over which smoothing of the data took places from 10 minutes to 30 minutes and finally to 2 hours acted to reduce the predicted background concentration in every sample. The results of the increased smoothing interval also reduced the magnitude of extreme values in the data, producing less scatter and a more precise relationship between predicted and measured background concentrations. Increasing the smoothing time period also resulted in the reduction of the slope of the relationship, resulting in lower accuracy in the predictions. Figure 7 shows the typical output for a smoothed minima background prediction.

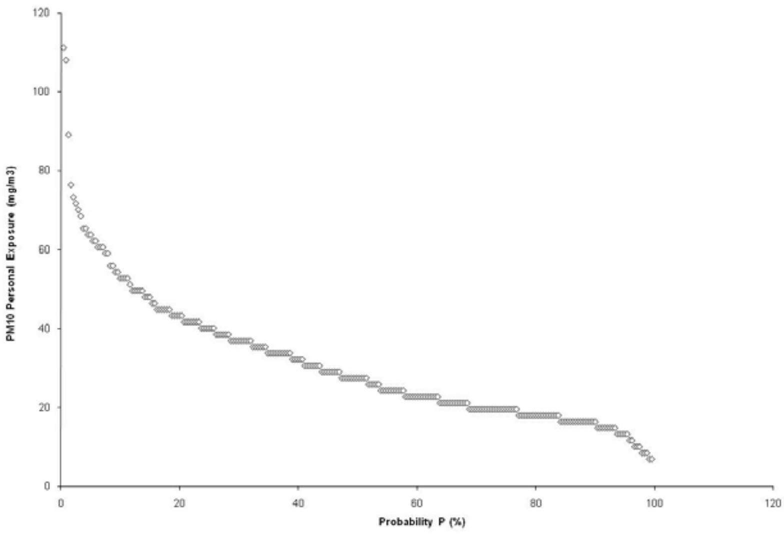

Figure 5. *Concentration-probability plot.*

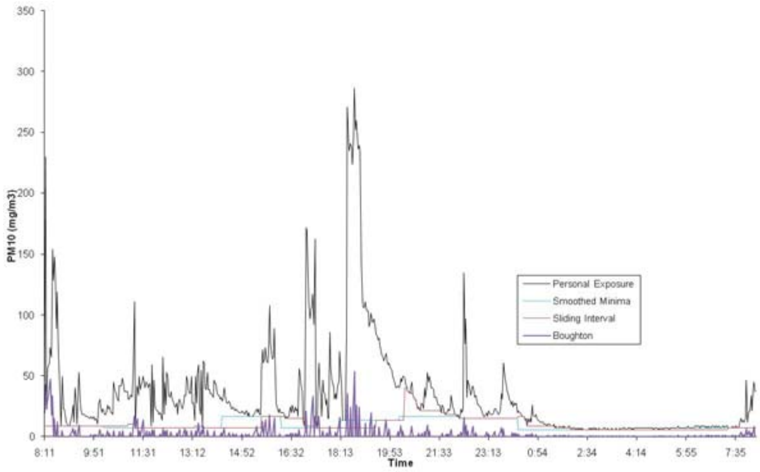

Figure 6. *Filter separation techniques: background concentration predictions from a 24-hr PM_{10} personal exposure time series plot.*

The best fit between predicted and measured data was achieved using a 0.5 hour smoothing interval. The precision of the predictions was found to be reasonably good with an R^2 of 49%, however this filter separation method underestimated the background concentrations with a slope of 0.61. The level of precision achieved was similar to that achieved using the Boughton method but the accuracy of prediction was significantly lower.

Sliding interval method. Using the sliding interval filter separation technique the personal exposure concentrations were also smoothed over a specified time period, however the difference between this and the previous technique was that the time period was overlapping. As a result the sliding interval separated background concentration produces a higher frequency time series to the previous method. This is evident when examining Figure 6. The sliding interval time period was initially chosen as 10 minutes which produced reasonably accurate but imprecise result as shown in Figure S2 (see the SM). These predictions were improved upon however by increasing the sliding interval to two hours. The precision of the sliding interval separated background predictions was again reasonably good at R^2 = 52%, similar to the smoothed minima technique. The accuracy of predictions was again poor however with a slope of 0.55, underestimating the measured background concentration. Overall the performance of the sliding interval is on a par with the smoothed minima method in terms of precision and marginally weaker than the smoothed minima method in terms of accuracy.

Recursive digital filters. Using RDF 1 to RDF 3 [Equations (1)–(3)] the RDF separated background concentrations were produced from the personal exposure data as shown in Figure 7. The recession constants in each of the equations were optimised to achieve a best fit against the measured data while maintaining realistic background time series profiles. For RDF 1 a recession constant of a = 0.4 was used, the resulting predictions were found to underestimate the measured background concentrations with a slope of 0.66. The precision of these predictions was however reasonably good with R^2 = 55% as shown in Figure S3 (see the SM). The performance of the RDF 1 equation was found to similar to that of the smoothed minima and sliding interval techniques.

For the second one–parameter algorithm RDF 2 a recession constant of k = 0.1 was also used which resulted in a lower frequency background time series. The accuracy and precision of the RDF 2 separated background predictions can be seen in Figure S3 (see the SM) whereby the measured background concentrations were underestimated with a slope of 0.49 on average and a precision of 49% was achieved. The use of RDF 2 therefore proved less useful than RDF 1.

The last RDF examined was a two–parameter algorithm and its recession constants were chosen for best fit as k = 0.2 and C = 0.4. Figure 7 shows a typical background time series separated from personal exposure data using RDF 3. It can be seen from Figure 7 that the frequency of predicted background concentrations is lower still using RDF 3. Comparing the predicted and measured data for RDF 3 reveals an underestimation of the measured background with a slope of 0.39 and a precision of 47%. Again despite the lower frequency response of RDF 3 compared to RDF 1 the use of this equation proved less useful in terms of accuracy and precision.

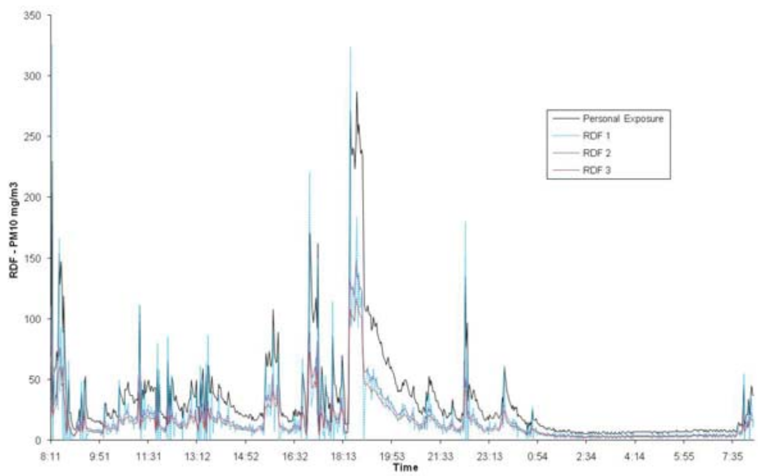

Figure 7. *RDF separated background concentration from a 24-hr PM_{10} personal exposure time series plot.*

3.2. Frequency analysis

The frequency analysis technique was examined for the accuracy and precision of its background concentration predictions, initially by assuming that the P_{50} personal exposure concentration was equal to the measured background concentration, i.e. that the personal exposure exceeded the background 50% of the time. The result of this analysis can be seen in Figure S4 (see the SM), however improvements on the initial performance were made by altering the assumed background concentration from the P_{50} value, the performance was instead investigated for P_{40}, P_{20} and P_{10}.

Choosing the personal exposure concentration at P_{40} for each sample as a measure of the background concentration produced the best fit results. The measured background concentrations were underestimated using this method with a reasonably high slope of 0.89. The precision of the predictions was however low in comparison to the results of previous techniques, with R^2 = 31%.

4. Discussion

Assessing the performance of each of the methods investigated shows variable results in the solution of this problem. Some methods produced favourable accuracy and/or precision but their predicted background concentration time series was, in some cases, of a higher frequency than the original personal exposure, contradicting the theory. Table 1 provides a summary of the performance of each of the methods considered.

Table 1. *Summary of background separation techniques performance – accuracy, precision, response*

Separation Technique	Accuracy	Precision (R^2, %)	Response Frequency
Boughton	0.99	60	High
Smoothed Minima	0.61	49	Low
Sliding interval	0.55	52	Low
RDF 1	0.66	55	High
Frequency Analysis (P_{40})	0.89	31	n/a

The Boughton method produced a prediction of background concentration comprising a series of spikes in response to increases and decreases in the personal exposure time series. The predicted data showed a good level of precision with an R^2 of 60% and a very good level of accuracy. The resulting background concentration time series did not reflect the expected low frequency "slow–response exposure", instead the predicted background time series had a similar frequency than the original personal exposure time series as shown in Figure 6. The amplitude of the response was however considerably lower than the personal exposure and as a result the Boughton method produced a low amplitude but high frequency response to instantaneous changes in local air quality. This prediction therefore does not truly reflect the theory that background is a low frequency response to changes in local air quality, instead it approximates the assumption.

The smoothed minima and sliding interval techniques both produced background time series plots of a low frequency, in line with the expected theory. The accuracy and precision of both was similar but both significantly underestimated the measured background concentrations and both had a precision less than that achieved using the Boughton method.

Using the RDF equations, the best performance was achieved using RDF 1, which resulted in a high frequency response. RDF2 and RDF3 produced much lower frequency responses in line with the theoretical assumptions but as a result, their predictions achieved less agreement with the measured data.

The frequency analysis method produced quite accurate results but had the weakest precision. In addition its output comprised a single value for background concentration as opposed the time series prediction given in the other methods, limiting its usefulness to a certain extent.

In all cases of this analysis the performance of the different techniques was influenced by the spatial and temporal resolution of the background concentration measurements to which they were being compared. It was assumed in the analysis that, in theory, the personal exposure of subjects can never fall below the background concentration of the environment in which they are exposed. However in the current study only one urban background monitoring station was available, located in an outdoor roadside environment. Clearly the background concentration of indoor office–type environments, in which subjects spent a significant portion of their time, could differ from the urban roadside concentration. In addition the background concentration was noted in Section 1 to change with time and the 24–hour average measurements provided may have given insufficient resolution for the optimum performance of the separation techniques presented. Both of these limitations in the resolution of the available background data may explain the low accuracy and/or precision of some of the techniques and it is clear that scope for improvement of this analysis method exists with the inclusion of higher resolution data. However, with these limitations in mind, the results of the present study are encouraging in terms of the potential of this methodology.

5. Conclusions

The evidence presented in this paper suggests that it is valid to assume that personal exposure is made up of various components of exposure including background concentration. It shows that it is possible to separate the background component from personal exposure data to a reasonable degree of accuracy and it also shows that synergies exist between the analysis techniques of air pollution and stream–flow hydrology.

The Boughton method has been demonstrated to provide the best results in background concentration separation in comparison to the other methods investigated. Future research in this area should aim to improve the accuracy and precision of background air pollution separation techniques, possibly through the inclusion of additional background monitoring locations in the analysis and at a higher temporal resolution.

This technique, although requiring further research to refine its performances and demonstrate its robustness, may provide the foundation for a valuable method of predicting personal exposure data from background concentrations or vice versa. Significant potential exists in this analysis technique to improve our ability to predict personal exposure to air pollution.

Acknowledgements

The authors would like to thank Environmental Protection Agency, STRIVE program for funding this investigation and Dublin City Council for providing data.

Supporting Material Available

Smoothed minima separated PM_{10} background concentrations vs. measured PM_{10} background concentrations – with varying non–overlapping smoothing time periods (10 to 120 min) (Figure S1), Sliding interval separated PM_{10} background concentrations vs. measured PM_{10} background concentrations–with varying sliding interval time periods (10 to 120 min) (Figure S2, RDF separated PM_{10} background concentrations vs. measured PM_{10} background concentrations (Figure S3), Frequency analysis separated background PM_{10} concentrations compared to measured background PM_{10} concentrations (Figure S4). This information is available free of charge via the Internet at http://www.atmospolres.com.

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