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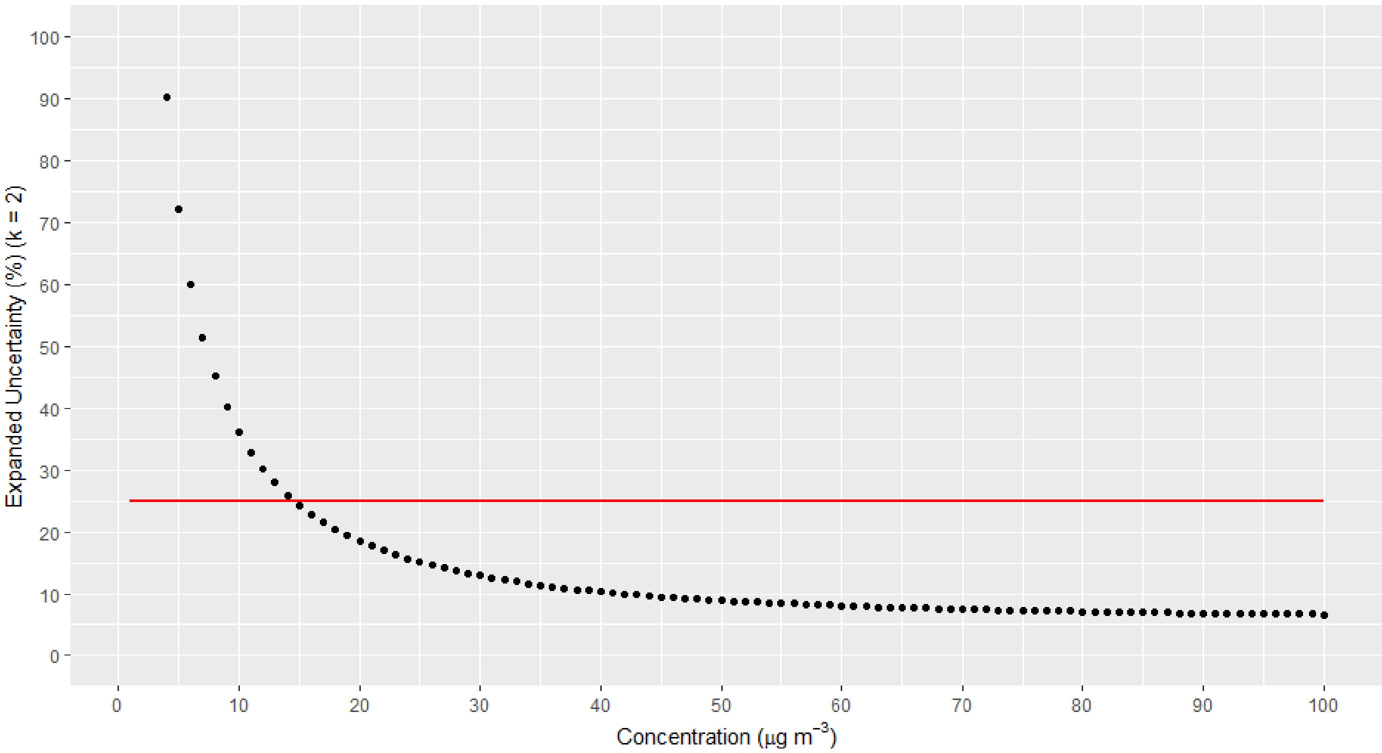

Pilot Research Study to Investigate Particulate Matter Monitoring Techniques in Scotland

Final Report

Report for Scottish Government

ED11195 | Issue 1 | Date 19/08/2021

Ricardo Confidential

Customer:

Scottish Government

Customer reference:

ED11195

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Ref: ED11195

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Executive summary

Ricardo Energy & Environment (Ricardo) were contracted by Scottish Government to investigate the relationship between automatic particulate matter (PM_{10} and $PM_{2.5}$) measurement techniques used in Scotland and the EU reference method. The requirement for a study of this type was identified following decreases in PM_{10} concentrations observed across Scotland's air quality monitoring network following the introduction of Fidas, which replaced TEOM, FDMS and BAM instruments. The reasons for this drop are complex with previous studies indicating that the Fidas (11) may not be correctly accounting for the mass adequately for particles smaller than 0.18 µm. Another factor that is likely to have contributed to this difference is the offset that can exist in FDMS and BAM data. Measured offsets of up to ±3 µg m^{-3} are not routinely corrected for due to the higher limits of detection (LoD) of these analysers, typically: FDMS LoD = ±5 µg m^{-3}; BAM LoD = ±6 µg m^{-3}. The performance of the Fidas at low concentrations is considerably better with the response to particle-free air is typically less than ±0.5 µg m^{-3}.

The aim of the study was to help identify the reason for a noticeable change in particulate matter (PM) concentrations; and also provide certainty in measured PM concentrations for authorities seeking to revoke PM_{10} air quality management areas (AQMAs). Due to the significant impact of particulate matter on human health, it is crucially important that PM_{10} AQMAs are not revoked unless it is certain that the objectives are not being exceeded. The initial pilot study was carried out over a 12-month period between January 2020 and January 2021.

As part of this study the following three measurement techniques were assessed against an EU reference method – MicroPNS Type HVS16 gravimetric sampler:

- Beta attenuation monitor (BAM) with heated inlet
- Tapered element oscillating microbalance – filter dynamics measurement system (TEOM-FDMS)
- Fidas with the current approved calculation algorithm for converting particle numbers to mass concentrations (Method 11) and an updated algorithm (Method 73), which is not currently approved for use.

The BAM, FDMS and Fidas (11) have all been assessed as equivalent to the reference method through the UK MCERTS scheme (https://uk-air.defra.gov.uk/networks/monitoring-methods?view=mcerts-scheme). The UK MCERTS scheme assessment identified that, for PM_{10} and $PM_{2.5}$ data to meet the equivalence criteria from the three automatic methods stated, the corrections for slope stated in the table below must be applied.

Analyser Type	Correction for Gravimetric Equivalence	
	PM_{10}	$PM_{2.5}$
Smart Heated BAM	Divide by 1.035	No correction
TEOM-FDMS	No correction	No correction
Fidas (11)	No correction	Divide by 1.060

The table below summarises the corrections for slope for gravimetric equivalence that were identified using the data from this study. It is important to emphasise that the tests undertaken for this study do not meet the requirements of a full equivalence test and as such, the results are only an indication of the possible correction required.

The table shows that the results differ significantly from that derived in the formal equivalence trials shown in the table above, with Fidas and FDMS analysers possibly under-reading PM_{10} concentrations by up to 22% and with the Fidas also under-reading $PM_{2.5}$ concentrations by up to 20%, compared to the reference method.

Instrument	Difference from Reference Method	
	PM_{10}	$PM_{2.5}$
BAM	No correction	No correction
FDMS	Divide by 0.888	No correction
Fidas (11)	Divide by 0.817	Divide by 0.808
Fidas (73)	Divide by 0.864	Divide by 0.821

The calculated measurement uncertainties for PM_{10} at the Limit Value of 50 $\mu g\ m^{-3}$ for the four automatic monitoring techniques before and after corrections for slope are provided in the table below. The uncertainty in BAM measurements is well below the threshold ±25% for equivalence with a measurement uncertainty of 7.3% at the Limit Value following corrections for offset during data ratification. For the remaining three techniques, none meet the ±25% threshold for equivalence without correction but all meet this requirement once corrected.

Instrument	PM_{10} Measurement Uncertainty at 50 $\mu g\ m^{-3}$	
	Before Correction for Slope	After Correction for Slope
BAM	±7.3%	No correction required
FDMS	±29.3%	±8.1%
Fidas (11)	±43.2%	±8.5%
Fidas (73)	±24.6%	±9.2%
Instrument	Before Correction for Offset	After Correction for Offset
BAM (raw)	±13.8	±7.3%

Details of the calculated measurement uncertainties for $PM_{2.5}$ at a Limit Value of 30 $\mu g\ m^{-3}$ for the four automatic monitoring techniques before and after corrections for slope are provided in the table below. The uncertainty in BAM measurements is below the threshold ±25% for equivalence with a measurement uncertainty of 17.8% at the Limit Value following corrections for offset during data ratification. The uncertainty in FDMS measurements is ±16.0% without correction for slope, which meets the requirement for reference equivalent data. For the remaining two Fidas techniques, neither meet the ±25% threshold for equivalence without correction for slope but both meet this requirement once corrected.

Instrument	$PM_{2.5}$ Measurement Uncertainty at 30 $\mu g\ m^{-3}$	
	Before Correction for Slope	After Correction for Slope
BAM	±17.8%	No correction required
FDMS	±16.0%	No correction required
Fidas (11)	±41.9%	±17.9%
Fidas (73)	±37.0%	±20.9%
Instrument	Before Correction for Offset	After Correction for Offset
BAM (raw)	±18.3	±17.8%

Measurement uncertainties at the Scottish Limit Values of 18 $\mu g\ m^{-3}$ for PM_{10} and 10 $\mu g\ m^{-3}$ (as a daily mean) have not been assessed up to now; and are detailed in the table below. These calculated uncertainties are significantly greater than that calculated at the EU Limit Values.

Instrument	PM_{10} Uncertainty at 18 $\mu g\ m^{-3}$ Daily Mean	$PM_{2.5}$ Uncertainty at 10 $\mu g\ m^{-3}$ Daily Mean
BAM	±19.7%	±56.0%
FDMS	±21.0%	±46.7%
Fidas (11)	±19.2%	±48.1%
Fidas (73)	±22.2%	±52.8%

Further analysis of measurement uncertainty in annual average concentrations was also carried out. The table below shows the average PM_{10} and $PM_{2.5}$ corrected concentrations measured between 16/01/2020 and 12/01/2021 and the associated uncertainties. As expected, the measurement uncertainties decrease when the daily datasets are averaged over the measurement period and range from ±2.6% to ±11.9% for PM_{10} and ±3.9% to ±17.6% for $PM_{2.5}$. The uncertainty in a daily mean of 15.1 $\mu g\ m^{-3}$ measured by the BAM, for example, is ±22.2% compared to ±2.6% average of all daily means.

Again, for completeness, BAM (raw) uncertainties have also been included. These result show that without correction for offset during the data ratification process the expanded relative measurement uncertainty in the annual PM_{10} and $PM_{2.5}$ means would be ±44.4% and ±23.6%, respectively.

Instrument	PM_{10} Average Concentration ($\mu g\ m^{-3}$)	PM_{10} Uncertainty	$PM_{2.5}$ Average Concentration ($\mu g\ m^{-3}$)	$PM_{2.5}$ Uncertainty
BAM	15.1	±2.6%	7.8	±11.6%
FDMS	13.6	±11.8%	6.7	±17.6%
Fidas (11)	14.0	±9.0%	8.4	±4.2%
Fidas (73)	15.5	±11.9%	8.6	±3.9%
MPNS	14.3	-	8.8	-
BAM (raw)	18.6	±44.4%	10.8	±23.6%

The results therefore indicate that current corrections for equivalence may not be accurately representing how the automatic monitoring methods respond at lower concentration levels and meteorological conditions, such as those observed in Scotland. In addition, though the data suggested that the BAM required no correction for slope, the study has demonstrated that carrying out baseline checks on a regular basis and correcting for identified offsets during the data ratification process is critical in reducing the measurement uncertainty in the daily and annual mean concentrations.

Taking into consideration the results of this study, Scottish Government decided to extend the study for a further 12 months. This extension will focus on the Fidas analyser and will be more closely aligned with EN 16450, using duplicate Fidas and MPNS samplers within the Glasgow Hope St monitoring site. This approach will provide a more robust dataset to determine whether it is appropriate to apply correction factors to Fidas data used with the Scottish Air Quality Database (SAQD) monitoring network

The following recommendations are also made:

- Local authorities using Fidas within the SAQD network should not consider revoking an AQMA for PM_{10} until the results and recommendations from the next stage of the study are published.
- For $PM_{2.5}$, annual mean concentrations of greater than 8 $\mu g\ m^{-3}$ using a Fidas might indicate that the annual mean objective of 10 $\mu g\ m^{-3}$ has been exceeded.
- Local authorities using FDMS within the SAQD network should only consider revoking an AQMA for PM_{10} if the measured annual mean is consistently 16 $\mu g\ m^{-3}$ or less.
- Given the significant impact on improving measurement uncertainty, it may be necessary to consider the implementation of routine baseline checks for BAM and FDMS analysers within the SAQD network. These results could then be applied as evidence in data ratification.

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1 Introduction

Ricardo Energy & Environment (Ricardo) were contracted by Scottish Government to investigate the relationship between automatic particulate matter (PM_{10} and $PM_{2.5}$) measurement techniques used in Scotland and the EU reference method. The aim of the study was to help identify the reason for a noticeable change in PM concentrations; and also provide certainty in measured PM concentrations for authorities seeking to revoke PM_{10} air quality management areas (AQMAs). Due to the significant impact of particulate matter on human health, it is crucially important that PM_{10} AQMAs are not revoked unless it is certain that the objectives are not being exceeded. This report provides the results from the 12-month ongoing equivalence research study carried out between January 2020 and January 2021, which compared automatic monitoring techniques used within the SAQD with the reference method in an environment more suited to provide data relevant to Scotland's climate.

1.1 Background

In recent years, trend analysis has identified a significant drop (2 – 4 $\mu g\ m^{-3}$) in PM_{10} concentrations across the SAQD network. This drop coincides with a change in measurement technique within the Scottish Air Quality Database (SAQD) network to Fidas as an alternative to beta attenuation monitors (BAM) and filter dynamics measurement systems (FDMS). Similar step changes have been seen with PM_{10} in the past, with the change from Tapered Element Oscillating Microbalance (TEOM) analysers to FDMS, for example. However, this step change brought concentrations well below the annual mean objective for many sites and has in turn prompted the move to seek revocation of PM_{10} AQMAs.

In addition, authorities and data users were also noting a discrepancy between model background maps (generated using FDMS rural background sites) and measured concentrations from roadside sites (measured using FIDAS). In some instances, measured Roadside site concentrations were found to be lower than background maps.

A previous study carried out by Kings College London, Bureau Veritas and Ricardo to assess the relationship between automatic PM measurements and reference method gravimetric samplers at several AURN locations in England and Wales locations found that:

- The relationship between SEQ[1] (reference method) and Partisol samplers is excellent.
- The relationship between BAM / FDMS / Fidas is relatively good. Daily average concentrations normally follow the trend BAM concentrations > FDMS concentrations > Fidas concentrations, but the relative differences between average measurements is small (2 - 3 $\mu g\ m^{-3}$ across the entire range). This is however, significant in terms of the Scottish Air Quality Objectives (Table 1).
- When looking at the hourly relationship between automatic analysers, there is a clear shift in the baseline of the Fidas compared to the FDMS (at roadside sites). The Fidas does not measure particles smaller than 180 nm, but instead uses an algorithm based on the particle size distribution to assess their contribution. It is possible that this algorithm underestimates the contribution when very close to traffic sources.
- Establishing the baseline for Fidas is considerably easier in data ratification than either the FDMS or BAM. The Fidas displays very little noise throughout the measurement range, whereas the signal noise in BAM and FDMS makes it difficult to identify the correct baseline. This may account for a large proportion of the 2-3 $\mu g\ m^{-3}$ difference seen with Fidas measurements.
- Research suggests that this apparent Fidas under-read is strongly correlated to black carbon and high nitrogen oxides (NOx) concentrations. This underread appears to be worst when the sampling inlet is less than 0.5 m from the kerb of a heavily trafficked road.

[1] https://www.et.co.uk/products/air-quality-monitoring/particulate-monitoring/seq-4750-sequential-gravimetric-sampler

Table 1 Particulate matter air quality objectives - Scotland

Pollutant	Air Quality Objective	
	Concentration	Measured as
$PM_{2.5}$	10 µg m^{-3}	annual mean
PM_{10}	50 µg m^{-3} not to be exceeded more than 7 times a year	24-hour mean
	18 µg m^{-3}	annual mean

Currently the SAQD incorporates BAM, FDMS, Tapered Element Oscillating Microbalance (TEOM) and Fidas for PM monitoring, all of which have been tested as equivalent to the reference method for measuring PM_{10} and $PM_{2.5}$ - Table 2 details the number of PM instruments by type. Full equivalence testing requires the use of two reference method and two identical analysers, operated over four discreet 40-day campaigns over two seasons at 2 different locations (160 days minimum), with strict requirements for the range of concentration measurements.

However, ongoing equivalence data currently available for the UK are based on measurements at background locations in London and Manchester; and the full equivalence study referred to above was also based at locations in the southern regions of the UK. The PM climate in Scotland is known to be significantly different to the south east of England with significantly lower concentrations. In addition, all equivalence studies currently evaluate measurement uncertainties at EU Limit and Target Values: to be better than 25% at 50 µg m^{-3} daily average for PM_{10}, and 25% at 30 µg m^{-3} daily average for $PM_{2.5}$. No investigation of analyser performance at the WHO recommended values, shown in Table 3, has been undertaken to date.

Table 2 Number of particulate matter instruments in the SAQD by instrument type

Instrument	Number of PM_{10} Instruments	Number of $PM_{2.5}$ Instruments
BAM	4	1
TEOM	1	1
FDMS	2	4
Fidas (11)	65	

Table 3 WHO air quality guidelines

Pollutant	Concentration	Measured as
$PM_{2.5}$	25 µg m^{-3} 10 µg m^{-3}	24-hour mean Annual mean
PM_{10}	50 µg m^{-3} 20 µg m^{-3}	24-hour mean Annual mean

As Scottish authorities move towards considering revoking PM_{10} air quality management areas (AQMA), it is essential that Scottish Government has as much certainty as possible in the data provided from analysers within the SAQD. To achieve this, Ricardo carried out a 12-month ongoing equivalence research study between January 2020 and January 2021, which compared automatic monitoring techniques used within the SAQD with the reference method in an environment more suited to provide data relevant to Scotland’s climate.

1.2 CEN Standard EN 16450

The CEN Standard EN 16450 – 'Automated measuring systems for the measurement of the concentration of particulate matter' sets out the testing regime for carrying out equivalence tests. These tests are undertaken in controlled conditions. CEN EN 16450 requires ongoing equivalence but does not provide guidance for corrective action if the ongoing assessment identifies an issue.

UK full equivalence tests were undertaken in two separate locations over four different exercises, using two reference samplers and two automatic analysers (minimum 160 days of concurrent measurements required from all four devices). These data are then used to calculate an averaged response for the candidate analyser and an associated measurement uncertainty. The measurement periods are carefully chosen to ensure a wide range of PM concentrations are measured, typically this means during spring and autumn, when volatile PM makes a significant contribution to PM concentrations.

In contrast, ongoing equivalence requirements are more relaxed; one reference and one automatic analyser, and no guidance is provided about how to deal with results that deviate from those obtained during full equivalence testing. This report therefore forms the beginning of a structured investigation of analyser performance and to determine what next steps are required in the investigation

It is important to note that this study does not fulfil the equivalence testing requirements of EN 16450 and is a more streamlined ongoing equivalence approach. However, the study does provide invaluable information regarding the ongoing equivalence status of automatic methods used within the SAQD network when measuring the lower PM concentrations typically experienced in Scotland.

2 Methodology

2.1 Measurement techniques investigated

As part of this study, the following three measurement techniques were assessed against an EU reference method that meets the requirements of EN 12341[2] (MicroPNS (MPNS) Type HVS16 gravimetric sampler):

- Beta attenuation monitor (BAM[3]) with smart-heated inlet.
- Tapered element oscillating microbalance – filter dynamics measurement system (TEOM-FDMS[4]).
- Fidas[5] with the current approved algorithm for converting particle numbers to mass concentrations (Method 11) and an updated algorithm (Method 73), which is not currently approved for use.

The Fidas Method 73 algorithm has been developed by the manufacturer to account for the under-estimation of particulate matter concentrations identified in previous studies.

As stated previously, the BAM, FDMS and Fidas (Method 11) have all been assessed as equivalent to the reference method, which was carried out through the UK MCERTS scheme (https://uk-air.defra.gov.uk/networks/monitoring-methods?view=mcerts-scheme). Table 4 details whether PM_{10} and $PM_{2.5}$ data from the three automatic methods require any correction for slope to meet the equivalence criteria.

Table 4 Corrections required for equivalence for PM_{10} and $PM_{2.5}$

Analyser Type	Correction for Gravimetric Equivalence	
	PM_{10}	$PM_{2.5}$
Smart Heated BAM	Divide by 1.035	No correction
TEOM-FDMS	No correction	No correction
Fidas (Method 11)	No correction	Divide by 1.060

[2] CEN Standard EN 12341:2014 - Ambient air. Standard gravimetric measurement method for the determination of the PM_{10} or $PM_{2.5}$ mass concentration of suspended particulate matter
[3] https://metone.com/products/bam-1020/
[4] https://www.thermofisher.com/order/catalog/product/1400AB#/1400AB
[5] https://www.palas.de/en/product/fidas200s

2.2 Sampling Regime

Monitoring of both PM_{10} and $PM_{2.5}$ is being carried out at Glasgow Kerbside (Hope St - http://www.scottishairquality.scot/latest/site-info.php?site_id=GLA4) between January 2020 and January 2021. This site was selected due to:

- The wide range of pollution concentrations historically measured at the site
- the site's proximity to a busy urban road.
- the size of the monitoring hut, which enabled the installation of the four samplers

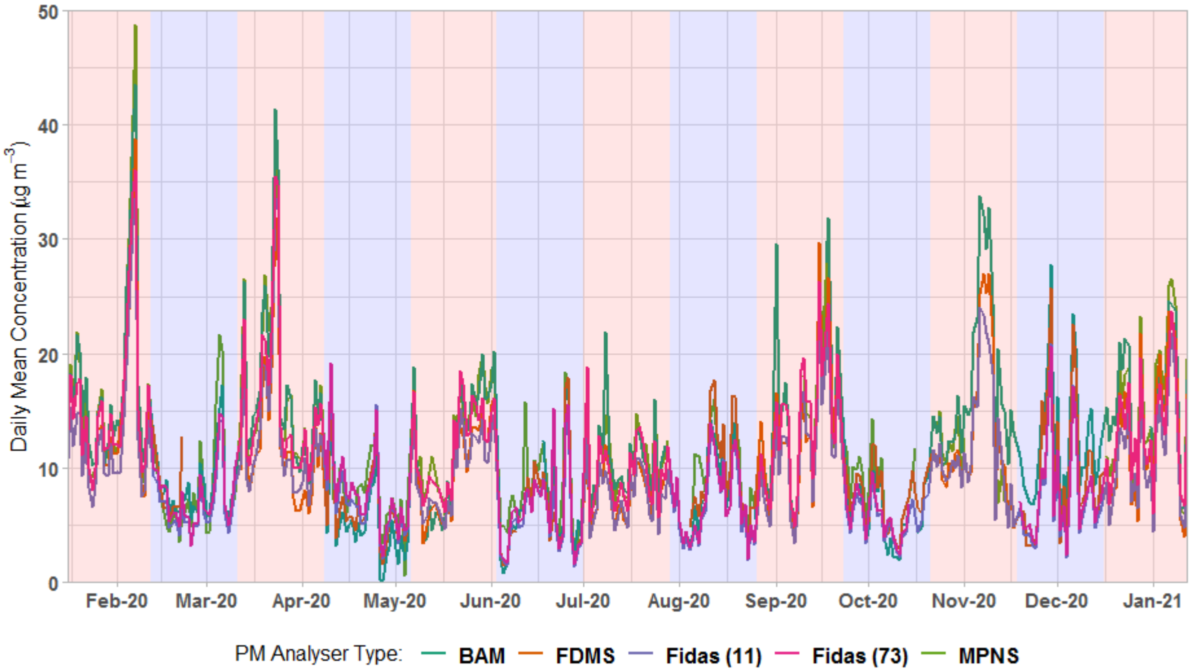
The Fidas analyser monitors both PM_{10} and $PM_{2.5}$ concentrations simultaneously. The conversion from particle counts to mass concentrations using both the Method 11 and Method 73 algorithms is carried out locally by the analyser. As a result, the Fidas provided four datasets: PM_{10} and $PM_{2.5}$ using Method 11, and PM_{10} and $PM_{2.5}$ using Method 73. Method 11 and Method 73 Fidas data are referred to as Fidas (11) and Fidas (73), respectively, from this point forward

Ideally, simultaneous monitoring of PM_{10} and $PM_{2.5}$ would have been carried out, however, this was not possible due to both the space restrictions within the existing monitoring hut and due to budget constraints. Therefore, for the BAM and FDMS, the analysers were switched between PM_{10} and $PM_{2.5}$ using the addition of a sharp cut-off cyclone (SCC) connected to the sample inlets; in combination with the PM_{10} sampling head. The MPNS PM_{10} sample head was swapped with a $PM_{2.5}$ head to measure the two size fractions. As a result, PM_{10} and $PM_{2.5}$ monitoring was carried out on alternate 4-weekly periods for these analysers. Figure 1 shows a time series plot of daily PM concentrations with the red shaded periods showing when PM_{10} was sampled and the blue, $PM_{2.5}$. The sampling schedule for PM_{10} and $PM_{2.5}$ is detailed in Table 5. Data were rejected from days where the samplers were swapped between PM_{10} and $PM_{2.5}$.

Table 5 PM_{10} and $PM_{2.5}$ sampling schedule

Pollutant Monitored	Start Date	End Date
PM_{10}	16/01/2020	12/02/2020
$PM_{2.5}$	12/02/2020	11/03/2020
PM_{10}	11/03/2020	08/04/2020
$PM_{2.5}$	08/04/2020	06/05/2020
PM_{10}	06/05/2020	03/06/2020
$PM_{2.5}$	03/06/2020	01/07/2020
PM_{10}	01/07/2020	29/07/2020
$PM_{2.5}$	29/07/2020	26/08/2020
PM_{10}	26/08/2020	23/09/2020
$PM_{2.5}$	23/09/2020	21/10/2020
PM_{10}	21/10/2020	18/11/2020
$PM_{2.5}$	16/12/2020	13/01/2021

Figure 1 Time series plot of daily mean particulate matter concentrations (shaded areas: Red = PM_{10}; Blue = $PM_{2.5}$)



2.3 QA/QC

There are a number of aspects of this study that require quality assurance/quality control:

- Sampler / analyser performance
- MPNS filter handling and weighing
- Data ratification

2.3.1 Sampler / analyser performance

In order to assess the performance of the MPNS sampler and automatic analysers, Ricardo carried out 6-monthly audits. Ricardo holds UKAS accreditation to ISO 17025 for flow rate checks on particulate (PM_{10} / $PM_{2.5}$) analysers and for the determination of the spring constant, k_0, for the TEOM analyser. ISO 17025 accreditation provides complete confidence that the analyser calibration factors are traceable to national metrology standards, that the calibration methods are sufficient and fit for purpose, and that the uncertainties are appropriate for data reporting purposes. The following instrument functional checks are undertaken at an audit:

- Leak and flow checks, to ensure that ambient air reaches the analysers, without being compromised in any way.

- TEOM k_0 evaluation. The factor is used to calculate particulate mass concentrations.

- Fidas verification check using calibration dust. If the Fidas does not measure the particle size correctly, this indicates that the data may need adjustment or rejection.

- Fidas zero check. This confirms that the measurements drop to zero when a filter is place on the sample inlet, if not, this indicates that the data may need adjustment or rejection

- Particulate analyser flowrates. Any error in the flow through these particulate analysers is directly reflected in an error in the final measure of particulate concentration.

- Assessing changes in local site environment. During the visit, a record of any changes in the site environment, for example any increase or decreased traffic flow due to road layout changes, construction activity, encroachment of the site by vegetation etc.

- Assessment of station infrastructure and operational procedures. Any deficiencies in site infrastructure or operational procedures, which may affect data quality or safe operation of the site, are noted.

In addition to the 6-monthly audits, baseline checks were carried out on the BAM and FDMS. These were carried out on an ad-hoc basis if a potential issue was identified during the daily data checks. Table 6 details the months where baseline checks were carried out of the BAM and FDMS. Two baseline checks of the FDMS were carried out and four of the BAM. An inconsistent offset of between +2.8 $\mu g\ m^{-3}$ and +4.8 $\mu g\ m^{-3}$ was identified in the BAM data on all four occasions and a baseline correction was applied to all BAM data as part of the data ratification process. No offset was identified in the FDMS data and so no correction has been applied.

Table 6 Automatic PM analyser baseline checks

Analyser	Month
BAM	January 2020
BAM and FDMS	August 2020
BAM	September 2020
BAM and FDMS	January 2021

2.3.2 MPNS filter handling and weighing

The MPNS sampler samples ambient air through a filter on a daily basis. The sampler holds a cartridge of 15 pre-weighed filters that are sampled over a two-week period and then reweighed. The total volume of ambient air sampled through each filter is recorded and the mass concentration is calculated using the change in weight of the filter and the volume of sampled air.

The following standard was adhered to in terms of the preparation, handling, sampling and weighing of the MPNS filters:

- EN12341 - Ambient air — Standard gravimetric measurement method for the determination of the PM_{10} or $PM_{2.5}$ mass concentration of suspended particulate matter.

2.3.3 Data ratification

When ratifying data, the following are closely examined:

- Issues that have been flagged up automatically by the software or during the daily checks
- zero and sensitivity factors used on each day – the baseline and flow checks feed into this
- General review of the result to make sure that there are no other anomalies.

Once the data have been initially ratified a proforma report is produced and passed to the data checker. The role of the data checker is to:

- Assess if there are any station problems if not the data can be marked as ratified.
- Return the station to the data ratifier if there are any issues requiring further action by the data ratifier.
- Forward the report for review by the wider project team if there are data quality issues which require a group discussion to resolve.

Following the final review, data are then adjusted if required and locked as ratified to the database. For this study, data are reviewed and processed on an ongoing basis, however, a final review of the full year's datasets is carried out prior to locking the data as ratified. The ratified datasets have then used in this report.

2.4 Data Analyses

2.4.1 Regression Analysis

Orthogonal regression analysis was used to investigate the relationships between the reference sampler and automatic analysers. The calculations used to carry out this analysis are detailed in Appendix 4, Section A4.1.

2.4.2 Identification of Outliers

The resultant regression model from the comparison of two datasets may consist of outliers and although these outliers are valid data, they may unduly influence the regression model. As a result, the identification of potential outliers was carried out using the generalised extreme studentised deviate (ESD) test[6]. Identified outliers were then rejected from the dataset and the regression analysis was carried out again.

3 Results

3.1 Monitoring Results

Average uncorrected PM_{10} and $PM_{2.5}$ concentrations measured between 16/01/2020 and 12/01/2021 are detailed in Table 7 and shown graphically in Figure 2 and 3, respectively. The results follow a similar pattern to what has been seen in previous studies, as discussed in the Introduction, where BAM PM_{10} concentrations > FDMS concentrations > FIDAS (11) concentrations. It is also seen that the difference is 3.7 µg m^{-3} across the whole range in this case.

The pattern is slightly different when looking at average $PM_{2.5}$ concentrations where both the BAM and FDMS measured 7.8 µg m^{-3} compared to 6.6 µg m^{-3} by the Fidas (11). However, if the range of $PM_{2.5}$ concentrations measured are taken into consideration, it can be seen in Figure 3 that again the same pattern is seen where BAM $PM_{2.5}$ concentrations > FDMS concentrations > FIDAS (11) concentrations.

As discussed in Section 2.1, the Fidas (73) algorithm has been developed to take account of the difference seen by Fidas (11). In this case, average PM_{10} and $PM_{2.5}$ concentrations measured by Fidas (73) are 2 and 0.5 µg m^{-3} greater than that measured by Fidas (11).

Table 7 Average uncorrected PM_{10} and $PM_{2.5}$ concentrations, 16/01/2020 – 12/01/2021

Instrument	Average PM_{10} Concentration (µg m^{-3})	Average $PM_{2.5}$ Concentration (µg m^{-3})
BAM	15.1	7.8
FDMS	12.1	7.8
Fidas (11)	11.4	6.6
Fidas (73)	13.4	7.1
MPNS	14.3	8.8

[6] Generalized ESD Test for Outliers: https://www.itl.nist.gov/div898/handbook/eda/section3/eda35h3.htm

Figure 2 Box plot of uncorrected daily mean PM_{10} concentrations, 16/01/2020 – 12/01/2021

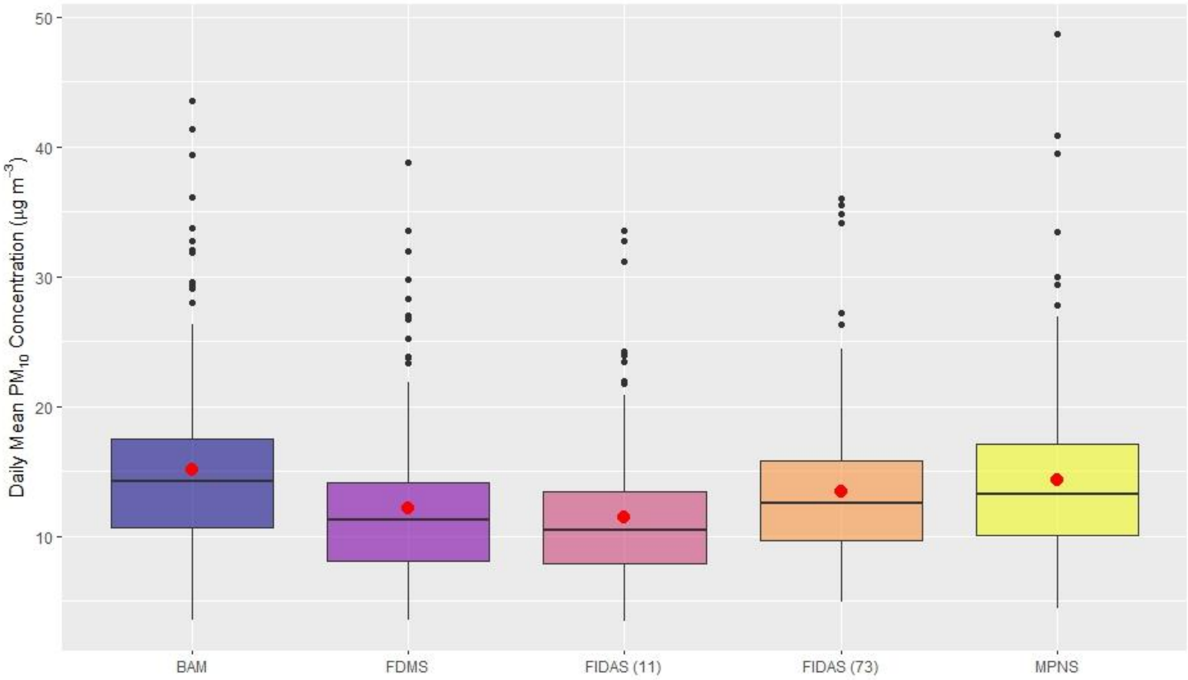
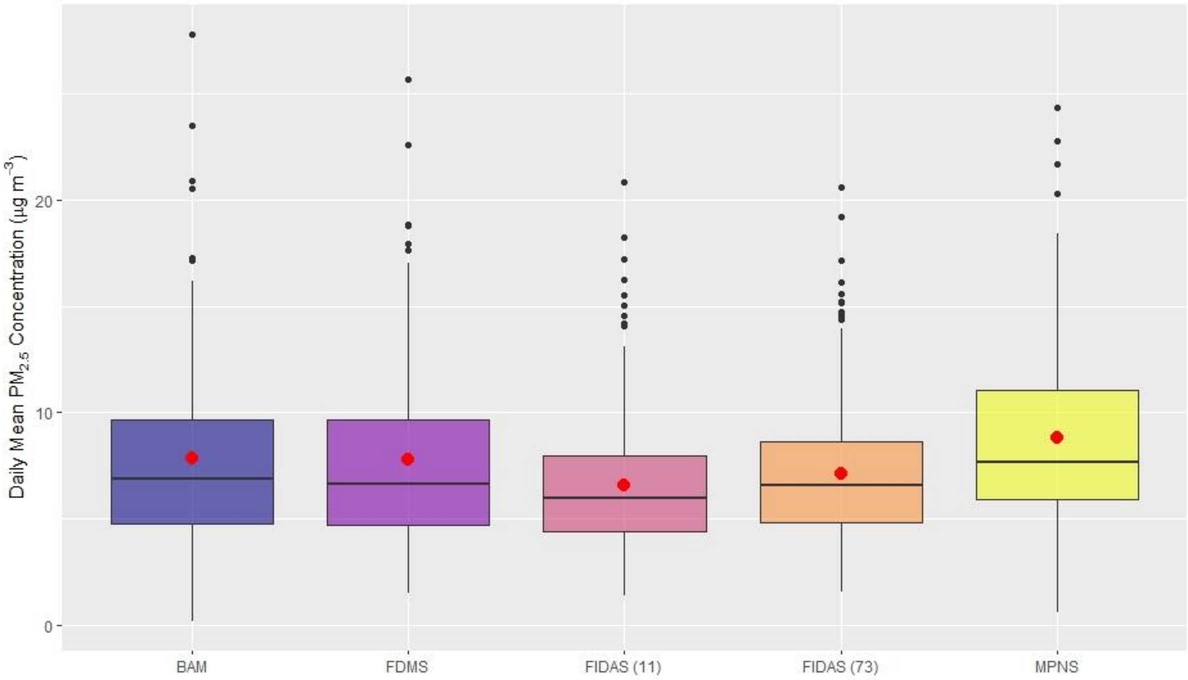


Figure 3 Box plot of uncorrected daily mean $PM_{2.5}$ concentrations, 16/01/2020 – 12/01/2021



3.1.1 Pollutant concentrations during 2020

The year 2020 was a unique year in terms of measured pollutant concentrations due to the Covid-19 pandemic and the lockdown restrictions that were in place as a result. Figures 4 to 6 show the NO_2, PM_{10} and $PM_{2.5}$ concentrations, respectively, averaged over all automatic monitoring sites in Glasgow as a time variation plot (http://www.scottishairquality.scot/data/openair?build=timevariation). These plots show how average pollutant concentrations vary by hour of the day, day of the week and month of the year for the years 2015 to 2020. The plots clearly show that significantly lower NO_2, PM_{10} and $PM_{2.5}$ concentrations were measured during 2020, compared to the five previous years.

A consequence of the lower concentrations during 2020 is that it becomes more difficult to assess the ongoing equivalence. This is especially apparent in the $PM_{2.5}$ results in Sections 3.2 and 3.3, where the noise of the instruments has a greater impact on the uncertainty of the final results. Ideally, a wide range of concentrations would be measured to enable better assessment of the analyser responses.

Figure 4 Average NO_2 concentrations measured at all sites in Glasgow, 2015 – 2020 ($\mu g\ m^{-3}$)

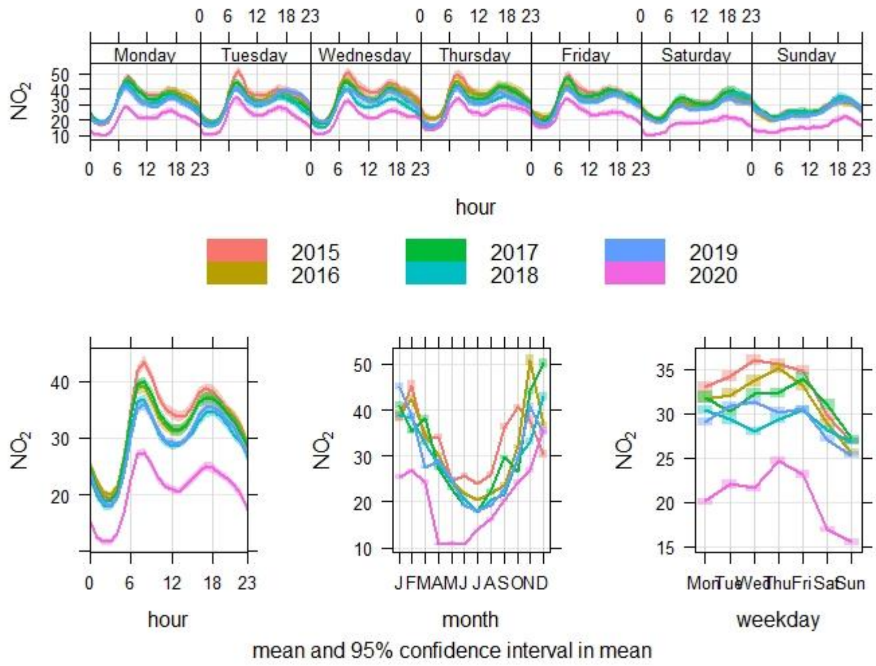


Figure 5 Average PM_{10} concentrations measured at all sites in Glasgow, 2015 – 2020 ($\mu g\ m^{-3}$)

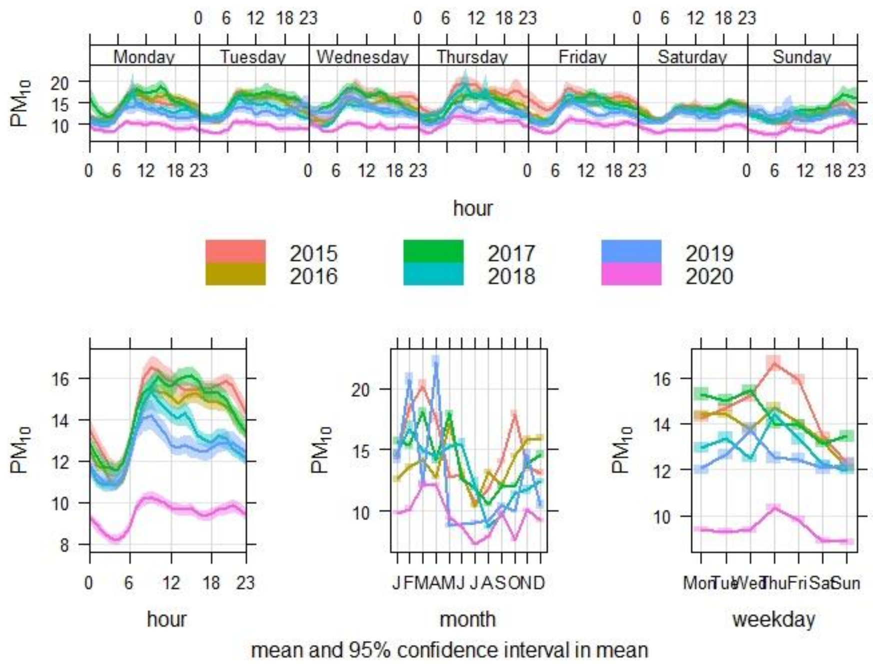
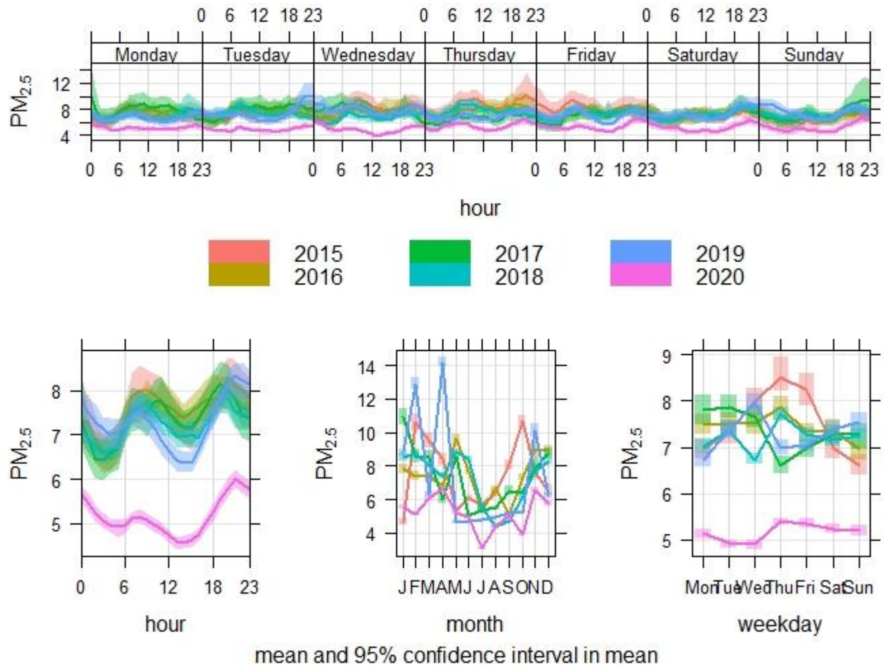


Figure 6 Average $PM_{2.5}$ concentrations measured at all sites in Glasgow, 2015 – 2020 (µg m^{-3})



3.2 Regression Results

For the study to be as robust as possible, the daily average concentration range needs to be as large as possible. EN 16450 states that it is a requirement for these types of studies to have 20% of measured daily average concentrations of PM_{10} and $PM_{2.5}$ above 28 µg m^{-3} and 17 µg m^{-3} respectively. During this study, six days of PM_{10} greater than 28 µg m^{-3} and seven days of $PM_{2.5}$ greater than 17 µg m^{-3} were measured. This limitation means that there is reduced confidence in the accuracy of the slope, offset and uncertainty results presented in this report.

Table 8 and Table 9 below detail the results from the regression analysis carried out for PM_{10} and $PM_{2.5}$, respectively, for the four automatic monitoring techniques. The number of outliers identified and removed from the datasets for the regression analysis are detailed. The significance columns indicate whether a correction for slope and/ or intercept is required using the criteria set out in Section A4.3 (Appendix 4). For example, the Fidas (11) PM_{10} results indicate that the slope is significant and so the data would need divided by 0.817 as the correction for equivalence.

It is important to re-emphasise that the regression analysis for the automatic techniques was carried out using data as would be ratified within the SAQD network. Specifically, for the BAM, corrections for offset identified during the QA/QC process were applied prior to the analysis. No additional corrections were applied or required for the Fidas and FDMS analysers. For completeness, Table 8 and

Table 9 also show the results for the un-corrected BAM data (BAM (raw)) and show that without correction, significant intercepts of +3.8 and +2.7 µg m^{-3} were identified for PM_{10} and $PM_{2.5}$, respectively.

The correlation of the PM_{10} automatic techniques is very strong for all techniques with R^2 of greater than 0.92. The data to date indicate that the BAM data do not require any correction for slope and offset to make the data equivalent to the reference data. However, the data indicate that FDMS and the Fidas (73) would need to be corrected for slope and intercept, and the Fidas (11) would need to be corrected for slope to make the measurements equivalent. All three methods are under-reading PM_{10} concentrations by between 13% and 22% when compared to the reference method. Regression scatter plots and results of uncorrected and corrected PM_{10} data for the four automatic monitoring techniques are shown in Appendix 1.

For $PM_{2.5}$, the correlation between the reference sampler and automatic techniques is weaker. This is likely due to the lower concentrations measured and the associated performance of the devices becoming more of a factor in the comparisons of the data. In this case, the data indicate that the BAM requires a correction for intercept, the FDMS for slope and intercept, and both Fidas methods require a correction for slope. The data indicate that the Fidas (11) and Fidas (73) under-read $PM_{2.5}$ by 24% and 22%, respectively, when compared to the reference method, and the FDMS over-reads by 12%. Regression scatter plots and results of uncorrected and corrected $PM_{2.5}$ data for the four automatic monitoring techniques are shown in Appendix 2.

Table 8 Orthogonal regression results for PM_{10}

Instrument	Number of Samples	No. Outliers	Orthogonal Regression								
			R^2	Slope (b) ± u_b			Significance	Intercept (a) ± u_a			Significance
BAM	165	4	0.92	1.009	±	0.022	Not Significant	0.1	±	0.4	Not Significant
FDMS	169	2	0.92	0.888	±	0.019	**Significant**	-1.1	±	0.3	**Significant**
Fidas (11)	178	2	0.92	0.817	±	0.017	**Significant**	-0.5	±	0.3	Not Significant
Fidas (73)	161	1	0.92	0.864	±	0.020	**Significant**	+0.8	±	0.3	**Significant**
BAM (raw)	165	5	0.93	0.984	±	0.021	Not Significant	+3.8	±	0.4	**Significant**

Table 9 Orthogonal regression results for $PM_{2.5}$

Instrument	Number of Samples	No. Outliers	Orthogonal Regression								
			R^2	Slope (b) ± u_b			Significance	Intercept (a) ± u_a			Significance
BAM	127	1	0.67	1.024	±	0.053	Not Significant	-1.2	±	0.5	**Significant**
FDMS	120	4	0.82	1.115	±	0.043	**Significant**	-1.8	±	0.4	**Significant**
Fidas (11)	138	1	0.71	0.808	±	0.039	**Significant**	-0.2	±	0.4	Not Significant
Fidas (73)	136	1	0.66	0.821	±	0.043	**Significant**	0.2	±	0.4	Not Significant
BAM (raw)	127	1	0.66	0.943	±	0.050	Not Significant	+2.7	±	0.5	**Significant**

Generally, requirements for zero corrections are ignored by TUV[7] (Technical Inspection Association) and the MCERTS[8] certification committee. This is mainly because it is very difficult to predict response consistently, especially for BAM and FDMS. As a result, only corrections for slope have been applied to ratified data for all methods where required. Table 10 summarises the corrections required for gravimetric equivalence and shows that the results differ significantly from that derived in the formal equivalence trials (shown in Table 4). It is important to stress that precision is the most important parameter in terms of the measurement data. These results do not imply poor precision but only that a correction for slope may be required. Correction factors for slope will be confirmed once further work has been undertaken to verify this initial ongoing equivalence assessment study.

[7] https://www.tuv.com/united-kingdom/en/
[8] https://uk-air.defra.gov.uk/networks/monitoring-methods?view=mcerts-scheme

Table 10 Corrections required for PM_{10} and $PM_{2.5}$ using the measurements collected between 16/01/2020 and 12/01/2021

Instrument	Difference from Reference Method	
	PM_{10}	$PM_{2.5}$
BAM	No correction	No correction
FDMS	Divide by 0.888	Divide by 1.115
Fidas (11)	Divide by 0.817	Divide by 0.808
Fidas (73)	Divide by 0.864	Divide by 0.821

3.3 Measurement Uncertainty

Measurement uncertainty is extremely important when reporting concentrations, both in demonstrating that the measurements are meeting the required data quality objectives and in providing context and confidence to a specific measurement. For example, if a daily average PM_{10} concentration of 55 $\mu g\ m^{-3}$ is measured but the uncertainty is ±100%, which can be typical using low-cost sensor technologies without appropriate QA/QC applied, then no conclusion can be made regarding whether an exceedance of the daily mean objective was actually measured. Conversely, if the uncertainty is ±7% then we can be confident that there was in fact an exceedance.

Table 11 details the calculated measurement uncertainties for PM_{10} at the Limit Value of 50 $\mu g\ m^{-3}$ for the four automatic monitoring techniques before and after corrections for slope, compared to the official equivalence results. Note that the letters defined within the brackets following the corrected uncertainties indicate whether the data were corrected for slope (S). The uncertainty in BAM measurements is well below the threshold ±25% for equivalence with a measurement uncertainty of 7.3% at the Limit Value with a correction for offset during data ratification. For the remaining three techniques, none meet the ±25% threshold for equivalence without correction but all meet this requirement once corrected.

Table 11 PM_{10} measurement uncertainty of daily average concentrations

Instrument	PM_{10} Measurement Uncertainty at 50 $\mu g\ m^{-3}$		
	Before Correction	After Correction	Official Equivalence Results
BAM	±7.3%	No correction	±9.3 (divided by 1.035)[9]
FDMS	±29.3%	±8.1% (S)	±8.7 (uncorrected)[10]
Fidas (11)	±43.2%	±8.5% (S)	±7.5% (uncorrected)[11]
Fidas (73)	±24.6%	±9.2% (S)	n/a

Table 12 details calculated measurement uncertainties for $PM_{2.5}$ at a Limit Value of 30 $\mu g\ m^{-3}$ for the four automatic monitoring techniques before and after corrections for slope, compared to the official equivalence results. The uncertainty in BAM measurements is ±17.8% without correction, which meets the requirement for reference equivalent data. The uncertainty in FDMS measurements is ±16.0% without correction for intercept and ±18.6% with a correction, both of which meet the requirement for equivalence. However, this would suggest that the FDMS $PM_{2.5}$ data should not be corrected for slope in this case.

For the remaining two Fidas techniques, neither meet the ±25% threshold for equivalence without correction for slope but both meet this requirement once corrected. Plots of PM_{10} and $PM_{2.5}$ measurement uncertainty as a function of measured daily concentrations for the four automatic

[9] UK Report on the Equivalence of the Smart Heated $_{PM10}$ BAM-1020, May 2014. Available online at: https://www.csagroupuk.org/wp-content/uploads/2015/05/Smart-BAM-1020-PM10-UK-Report-with-manual-Final.pdf

[10] UK Equivalence Programme for Monitoring of Particulate Matter, June 2006. Available online at: https://uk-air.defra.gov.uk/assets/documents/reports/cat05/0606130952_UKPMEquivalence.pdf

[11] UK Report on the Equivalence of the Palas Fidas 200 Method 11 for PM_{10} and $PM_{2.5}$, March 2016. Available online at: https://www.csagroupuk.org/wp-content/uploads/2016/04/Palas-UK-Report-Final-with-Manuals-080316.pdf

monitoring techniques are shown in Appendix 3. The calculations used for the regression and measurement uncertainty analysis are shown in Appendix 4.

Table 12 $PM_{2.5}$ measurement uncertainty of daily average concentrations

Instrument	$PM_{2.5}$ Measurement Uncertainty at 30 µg m^{-3}		
	Before Correction	After Correction	Official Equivalence Results
BAM	±17.8%	No correction required	±12.6% (uncorrected)[12]
FDMS	±16.0%	**±18.6%** (S)	±16.4% (uncorrected at 35 µg m^{-3})[13]
Fidas (11)	±41.9%	±17.9% (S)	±9.3% (divided by 1.06)[14]
Fidas (73)	±37.0%	±20.9% (S)	n/a

One of the objectives of this study is to investigate measurement uncertainty as a function of mass concentrations. As described above, PM_{10} and $PM_{2.5}$ measurement uncertainties are calculated at specified Limit Values: 50 µg m^{-3} and 30 µg m^{-3} as daily averages, respectively. These Limit Values are much greater than the annual mean air quality objectives of 18 µg m^{-3} and 10 µg m^{-3} for PM_{10} and $PM_{2.5}$, respectively (Table 1). As a result, it is important to investigate how the measurement changes at these lower Limit Values.

Table 13 details the calculated measurement uncertainties for the four automatic analysers using the Scottish PM_{10} and $PM_{2.5}$ annual mean objectives as the Limit Values. All the analysers achieve measurement uncertainties of less than ±25% for PM_{10} at 18 µg m^{-3}. For $PM_{2.5}$ however, the measurement uncertainties range from ±46.7% (FDMS) to ±56.0% (BAM) at 10 µg m^{-3}, well above ±25%. This demonstrates that as PM concentrations decrease the measurement uncertainty in the daily average concentrations increases significantly.

Table 13 Measurement uncertainty at daily average Limit Values equal to the Scottish annual mean objectives

Analyser Type	PM_{10} Uncertainty at 18 µg m^{-3}	$PM_{2.5}$ Uncertainty at 10 µg m^{-3}
BAM	±19.7%	±56.0%
FDMS	±21.0%	±46.7%
Fidas (11)	±19.2%	±48.1%
Fidas (73)	±22.2%	±52.8%

Further analysis of the PM_{10} and $PM_{2.5}$ measurement uncertainties is shown in Table 14. In this case, the daily mean concentrations at which the measurement uncertainty becomes greater than ±25% are detailed. For PM_{10}, the results indicate that the uncertainty remains less than ±25% well below 18 µg m^{-3} for all four automatic methods. This together with the results in Table 13 would suggest that for a calculated annual mean of 18 µg m^{-3} using these methods, the uncertainty will be significantly less than ±25%.

The results are more uncertain for $PM_{2.5}$; they indicate that the uncertainty in the daily mean increases above ±25% at concentrations of between 19 µg m^{-3} for the Fidas (11) and 23 µg m^{-3} for the Fidas(73). Therefore, if comparing $PM_{2.5}$ data to the WHO daily mean guideline of 25 µg m^{-3}, the results indicate that uncertainty remains below ±25% for all methods. The provisional results also indicate that the uncertainty in annual mean $PM_{2.5}$ concentrations will likely be greater than that for PM_{10}.

[12] Certification Report and Checklist on the Evaluation of the Ambient Air Particulate Matter Test Reports Submitted for Approval and Certification within the MCERTS Scheme for UK Particulate Matter, October 2013. Available online at: https://www.csagroupuk.org/wp-content/uploads/2015/05/MCERTSCCPMT2BAMMet1013.pdf

[13] UK Equivalence Programme for Monitoring of Particulate Matter, June 2006. Available online at: https://uk-air.defra.gov.uk/assets/documents/reports/cat05/0606130952_UKPMEquivalence.pdf

[14] UK Report on the Equivalence of the Palas Fidas 200 Method 11 for PM_{10} and $PM_{2.5}$, March 2016. Available online at: https://www.csagroupuk.org/wp-content/uploads/2016/04/Palas-UK-Report-Final-with-Manuals-080316.pdf

Table 14 PM concentrations at which measurement uncertainty becomes greater than ±25%

Analyser Type	Daily PM_{10} Concentration at which Uncertainty > ±25% ($\mu g\ m^{-3}$)	Daily $PM_{2.5}$ Concentration at which Uncertainty > ±25% ($\mu g\ m^{-3}$)
BAM	14	21
FDMS	15	20
Fidas (11)	13	19
Fidas (73)	15	23

It is also appropriate to look at how the measurement uncertainty of daily average measurements impacts on longer average data e.g. annual means. The method used to calculate the measurement uncertainty in the calculated mean of PM_{10} and $PM_{2.5}$ concentrations is provided in Section A4.8, Appendix 4. It is important to note that there is no standard method for carrying out this calculation and so the process used in this report only provides an indication of the uncertainty in the calculated mean concentrations.

Table 15 shows the average PM_{10} and $PM_{2.5}$ corrected concentrations measured between 16/01/2020 and 12/01/2021 and the associated uncertainties. As expected, the measurement uncertainties decrease when the daily datasets are averaged over the measurement period and range from ±2.6% to ±11.9% for PM_{10} and ±3.9% to ±17.6% for $PM_{2.5}$. The uncertainty in a daily mean of 15.1 $\mu g\ m^{-3}$ measured by the BAM, for example, is ±22.2% compared to ±2.6% average of all daily means.

Table 15 Average corrected PM_{10} and $PM_{2.5}$ concentrations and associated measurement uncertainty

Analyser Type	PM_{10}		$PM_{2.5}$	
	Average Concentration ($\mu g\ m^{-3}$)	Uncertainty	Average Concentration ($\mu g\ m^{-3}$)	Uncertainty
BAM	15.1	±2.6%	7.8	±11.6%
FDMS	13.6	±11.8%	6.7	±17.6%
Fidas (11)	14.0	±9.0%	8.4	±4.2%
Fidas (73)	15.5	±11.9%	8.6	±3.9%
MPNS	14.3	-	8.8	-

Looking at the uncertainty calculations in more detail, there are two components; firstly, the random term which cannot be corrected for; and secondly bias which could be corrected for if it can be measured / characterised accurately (Table 16). In this case, the random uncertainty is quite consistent between the analyser types. The bias, however, manifests itself as a potential offset in the measurement data and has a greater influence on the final uncertainty. The raw BAM uncorrected data have also been included to demonstrate the impact that offsets can have on the measurement uncertainty.

As discussed in previous sections, the BAM PM_{10} and $PM_{2.5}$ data were corrected for offsets in the data and so this has reduced the bias in the BAM data and in turn reduced the uncertainty in the mean. No correction for offset was applied to any other analyser types as the zero checks did not highlight a significant offset. As a result, for PM_{10} specifically, we see that the uncertainty in the mean concentrations for the FDMS and Fidas is significantly greater than that calculated for the BAM. Similarly, for $PM_{2.5}$, the bias in the mean concentrations is greater in the BAM and FDMS, compared to the two Fidas methods, and so the uncertainty in the means is greater.

Table 16 Uncertainty random term and bias at calculated annual mean PM_{10} and $PM_{2.5}$ concentrations

Analyser Type	PM_{10} Random Term ($\mu g\ m^{-3}$)	PM_{10} Bias ($\mu g\ m^{-3}$)	$PM_{2.5}$ Random Term ($\mu g\ m^{-3}$)	$PM_{2.5}$ Bias ($\mu g\ m^{-3}$)
BAM (offset corrected during data ratification)	1.758	0.201	2.624	-1.026
FDMS (corrected for slope)	1.603	0.997	1.604	-1.636
Fidas (11) (corrected for slope)	1.599	-0.673	2.380	-0.334
Fidas (73) (corrected for slope)	1.750	0.928	2.616	0.267
BAM (no correction for offset)	2.305	3.858	2.843	2.312

Table 17 details the calculated measurement uncertainties of BAM PM_{10} and $PM_{2.5}$, using data that were not corrected for offset during the data ratification process. Although the uncertainties at the Limit Values are still below the ±25% objective, they are significantly greater than shown in Table 11 and Table 12. Correcting the offsets in the BAM data during data ratification reduced the calculated measurement uncertainties in PM_{10} concentrations by ±6.5% and ±0.5, respectively. A more dramatic impact is seen in the calculated uncertainties in the annual PM_{10} and $PM_{2.5}$ means with calculated uncertainties of ±44.4% and ±23.6% compared to ±2.6% and ±11.6% when corrected for offset during ratification. This highlights the importance of being able to identify and correct for offsets in BAM PM measurement data for improving the data quality. It is important to note that the reference MPNS sampler may also have a bias which has not been investigated within this study and would also have an impact on the uncertainty calculations.

Table 17 Calculate PM_{10} and $PM_{2.5}$ measurement uncertainties for BAM data not corrected for offset during data ratification

BAM not corrected for offset during data ratification	
PM_{10} Measurement Uncertainty at the Limit Value of 50 $\mu g\ m^{-3}$	±13.8%
$PM_{2.5}$ Measurement Uncertainty at the Limit Value of 30 $\mu g\ m^{-3}$	±18.3%
Uncertainty in annual average PM_{10} of 18.6 $\mu g\ m^{-3}$	±44.4%
Uncertainty in annual average $PM_{2.5}$ of 10.8 $\mu g\ m^{-3}$	±23.6%

4 Discussion and Conclusions

Ricardo Energy & Environment (Ricardo) were contracted by Scottish Government to investigate the relationship between automatic particulate matter (PM_{10} and $PM_{2.5}$) measurement techniques used in Scotland and the EU reference method. The requirement for a study of this type was identified following decreases in PM_{10} concentrations observed across Scotland's air quality monitoring network following the introduction of Fidas, which replaced TEOM, FDMS and BAM instruments. The reasons for this drop are complex with previous studies indicating that the Fidas (11) may not be correctly accounting for the mass adequately for particles smaller than 0.18 µm. Another factor that is likely to have contributed to this difference is the offset that can exist in FDMS and BAM data. Measured offsets of up to ±3 $\mu g\ m^{-3}$ are not routinely corrected for due to the higher limits of detection (LoD) of these analysers, typically: FDMS LoD = ±5 $\mu g\ m^{-3}$; BAM LoD = ±6 $\mu g\ m^{-3}$. The performance of the Fidas at low concentrations is considerably better with the response to particle-free air is typically less than ±0.5 $\mu g\ m^{-3}$. As a result, and as also shown in this study, the offsets are likely to be a contributor to the difference in PM_{10} concentrations seen at monitoring sites in the SAQD with the switch to Fidas (11).

It is important to note that these results do not supersede the equivalence designation for the three automatic methods but do highlight that ongoing equivalence testing needs to be carried out. The results also highlight that current corrections for equivalence may not be accurately representing how the automatic monitoring methods respond in Scotland's pollution and meteorological environment.

The results indicate that all automatic methods can meet the ±25% equivalence requirement easily for PM_{10} at the 50 µg m^{-3} daily average Limit Value. Calculated uncertainties for $PM_{2.5}$ conform to the requirements but are all significantly closer to 25% at the 30 µg m^{-3} daily average Limit Value. Table 18 summarises the correction factors derived from this study to achieve this requirement using the data collected between 16/01/2020 and 12/01/2021.

The latest Fidas (73) algorithm has not been assessed for equivalence and is not currently used within UK monitoring networks. The data indicate that this algorithm does correct somewhat for the difference seen using the current Fidas (11) algorithm. However, this study indicates that a significant correction for slope may still be required for both PM_{10} and $PM_{2.5}$ for Fidas (73) data.

The data indicate that the Fidas (11) under-reads by 22% and 24% for PM_{10} and $PM_{2.5}$, respectively, compared to the reference method. This suggests that FIDAS within the SAQD network measuring concentrations of 14 µg m^{-3} or greater could be exceeding the annual mean objective of 18 µg m^{-3}. For $PM_{2.5}$, measured annual mean concentrations of greater than 8 µg m^{-3} might indicate that the annual mean objective of 10 µg m^{-3} has been exceeded.

For the smart-heated BAM, the data indicate that no correction is required for PM_{10} and $PM_{2.5}$ (after bias correction for baseline offset). However, it has been demonstrated that carrying out baseline checks on a regular basis and correcting for identified offsets is critical in reducing the measurement uncertainty in the daily and annual mean concentrations.

The data indicate that the FDMS requires a correction for slope for PM_{10}, which under-read concentrations by approximately 12.5%. This indicates FDMS within the SAQD network measuring concentrations of 16 µg m^{-3} or greater could be exceeding the annual mean objective of 18 µg m^{-3}. The data indicate that no correction of FDMS $PM_{2.5}$ is required but again, regular baseline checks and adjustment for offsets are important for FDMS in order to minimise the uncertainty in the annual mean.

Table 18 Corrections required for PM_{10} and $PM_{2.5}$, 2020

Instrument	Difference from Reference Method	
	PM_{10}	$PM_{2.5}$
BAM	No correction	No correction
FDMS	Divide by 0.888	No correction
Fidas (11)	Divide by 0.817	Divide by 0.808
Fidas (73)	Divide by 0.864	Divide by 0.821

Taking into consideration the results of this study, Scottish Government have decided to extend the study for a further 12 months. This extension will focus on the Fidas analyser and will be more closely aligned with EN 16450, using duplicate Fidas and MPNS samplers within the Glasgow Hope St monitoring site. This approach will provide a more robust dataset to determine whether it is appropriate to apply correction factors to Fidas data used with the SAQD monitoring network

The following recommendations are also made:

- Local authorities using Fidas within the SAQD network should not consider revoking an AQMA for PM_{10} until the results and recommendations from the next stage of the study are published.

- For $PM_{2.5}$, annual mean concentrations of greater than 8 µg m^{-3} using a Fidas might indicate that the annual mean objective of 10 µg m^{-3} has been exceeded.

- Local authorities using FDMS within the SAQD network should only consider revoking an AQMA for PM_{10} if the measured annual mean is consistently 16 $\mu g\ m^{-3}$ or less.

- Given the significant impact on improving measurement uncertainty, it may be necessary to consider the implementation of routine baseline checks for BAM and FDMS analysers within the SAQD network. These results could then be applied as evidence in data ratification.

Appendices

A1 PM_{10} Regression Plots

Figures 7 to 17 show the PM_{10} orthogonal regression plots for the comparison between the MPNS reference sampler and the four automatic methods. The automatic data have been corrected based on the significance results detailed in Table 2. Plots of both uncorrected and corrected data are shown – in this case BAM PM_{10} data did not require any correction and so only uncorrected data are shown (Figure 7).

Two lines are shown in the plots:

- The black 1:1 line.
- The red orthogonal regression line.

For the plots with outliers removed, the identified outliers are shown as red triangles – see Figure 8 as an example. A summary of the regression analysis is shown in Tables 18 – 21.

Figure 7 Orthogonal regression plot – all BAM PM_{10} data (corrected for offset)

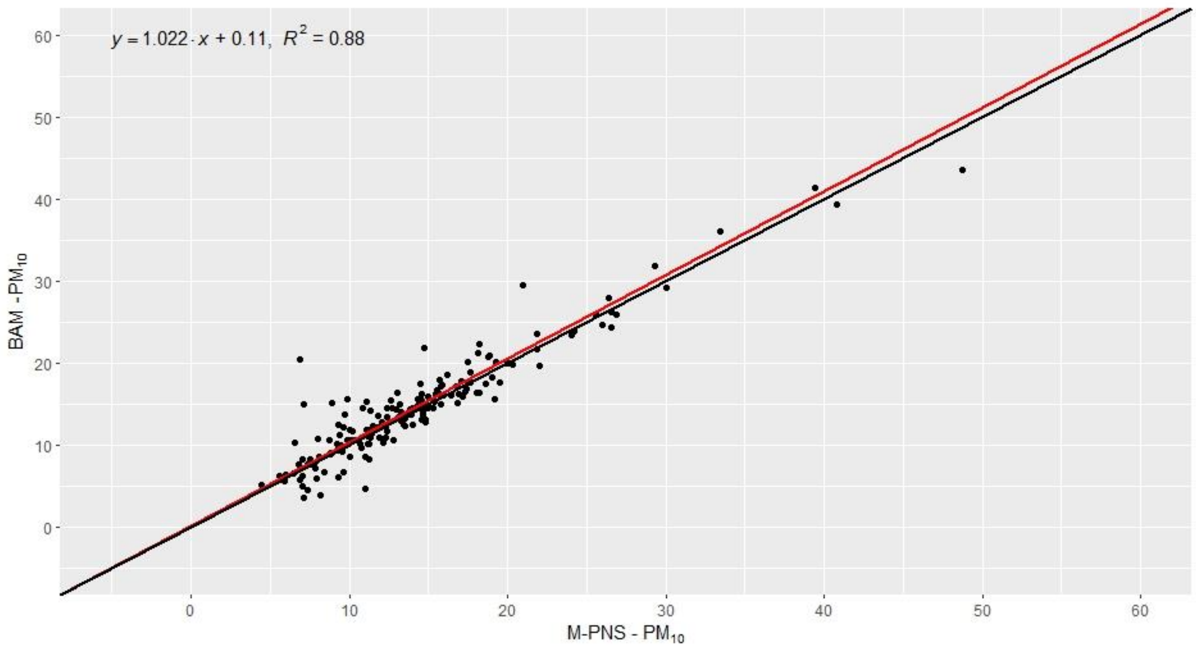


Figure 8 Orthogonal regression plot – BAM PM_{10} data (corrected for offset) with four outliers removed

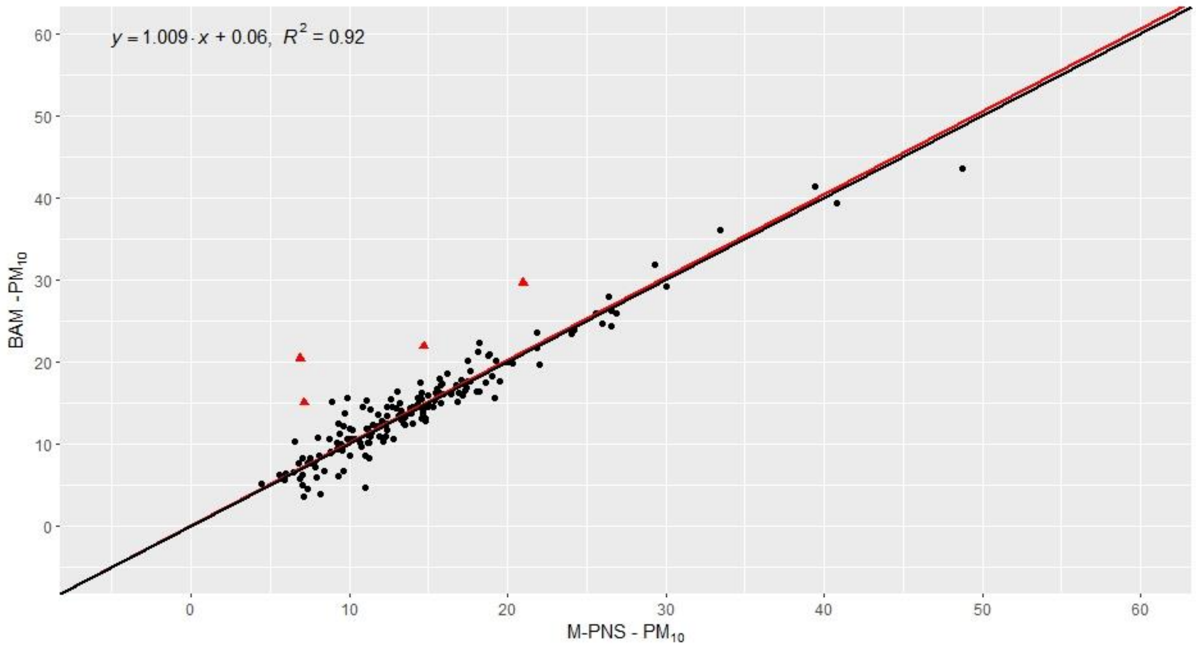


Table 19 Summary results – BAM PM_{10}

Measure	Uncorrected Data	Uncorrected Data - Outliers Removed
Intercept	0.108 ± 0.434 (Not Significant)	0.06 ± 0.35 (Not Significant)
Slope	1.022 ± 0.028 (Not Significant)	1.009 ± 0.022 (Not Significant)
Expanded Uncertainty at 50 µg m^{-3} (%)	10.301 (Pass)	7.347 (Pass)
R^2	0.88	0.923
Number of Measurements	165	161
Root Mean Square Error (µg m^{-3})	2.4	1.9
Mean Absolute Error (µg m^{-3})	1.6	1.4

Figure 9 Orthogonal regression plot – uncorrected BAM PM_{10} data with four outliers removed

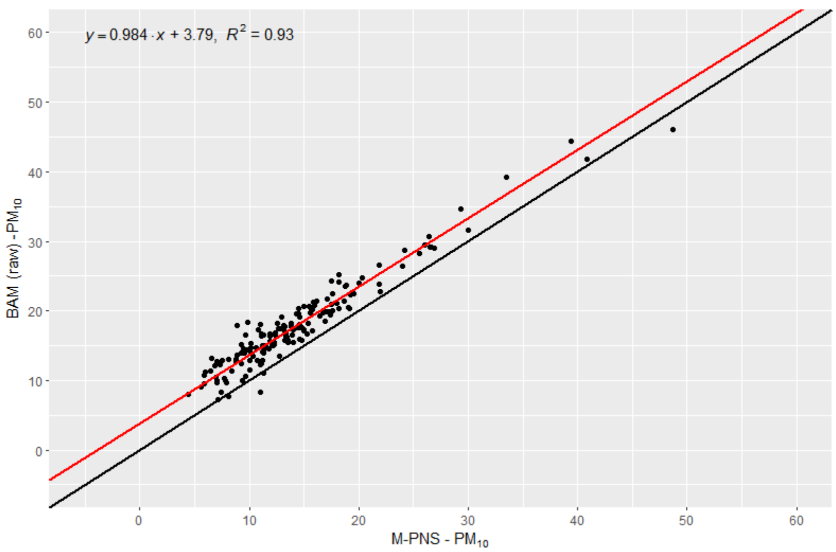


Figure 10 Orthogonal regression plot – all uncorrected FDMS PM_{10} data

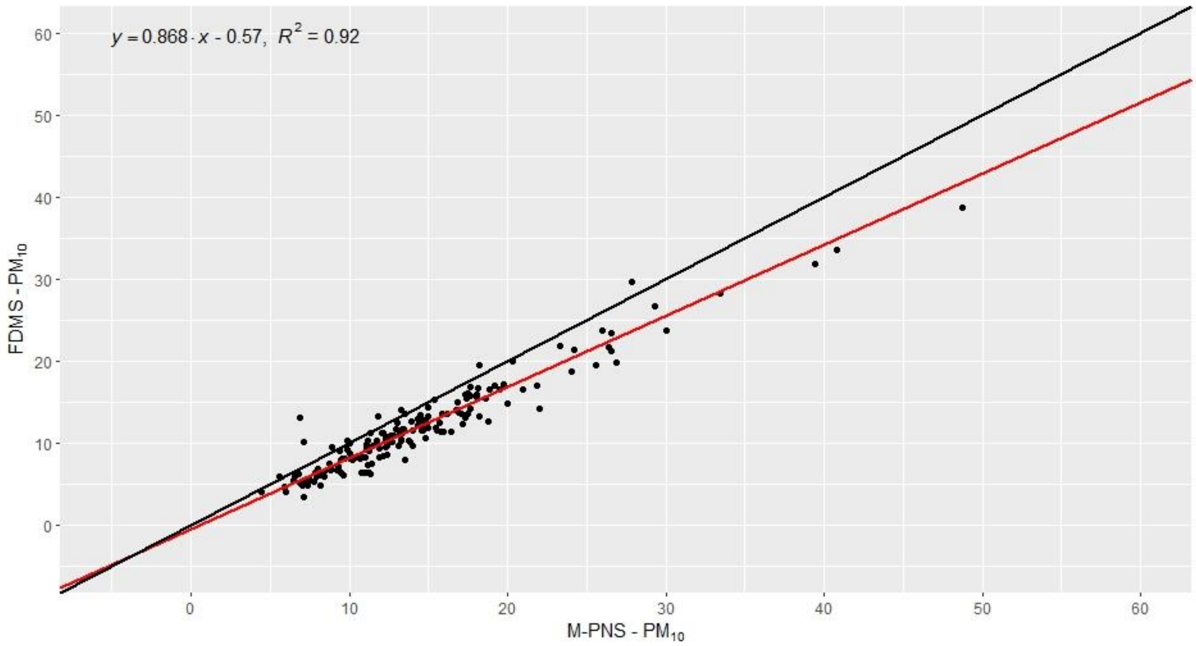


Figure 11 Orthogonal regression plot – uncorrected FDMS PM_{10} data with two outliers removed

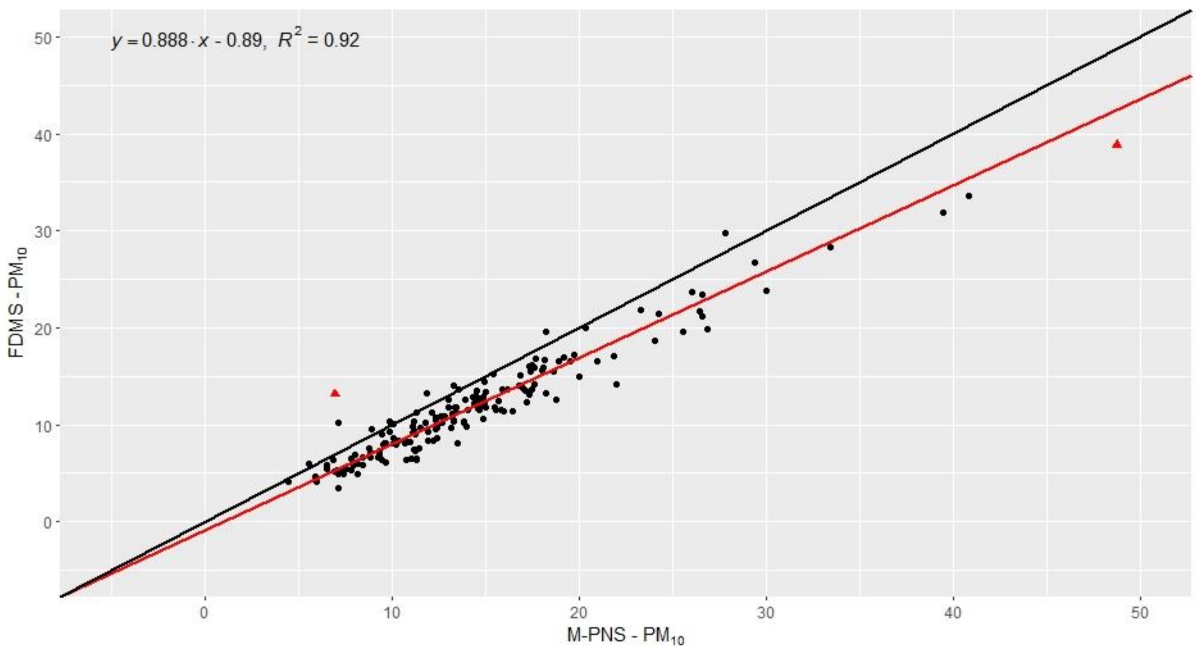


Figure 12 Orthogonal regression plot – FDMS PM_{10} divided by 0.888

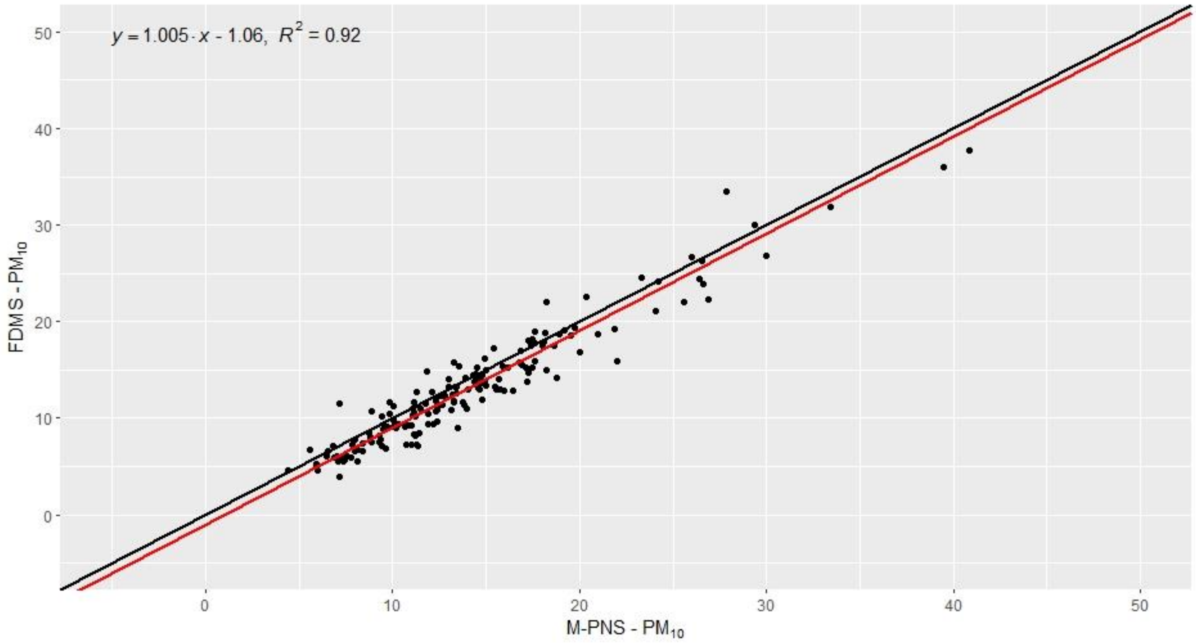


Table 20 Summary results – FDMS PM_{10}

Measure	Uncorrected Data	Corrected Data (Divided by 0.868)	Uncorrected Data - Outliers Removed	Corrected Data - Outliers Removed (Divided by 0.888)
Intercept	-0.569 ± 0.296 (Not Significant)	-0.739 ± 0.341 (Significant)	-0.886 ± 0.295 (Significant)	-1.064 ± 0.332 (Significant)
Slope	0.868 ± 0.019 (Significant)	1.006 ± 0.022 (Not Significant)	0.888 ± 0.019 (Significant)	1.005 ± 0.022 (Not Significant)
Expanded Uncertainty at 50 µg m^{-3} (%)	29.3 (Fail)	8.2 (Pass)	26.4 (Fail)	8.1 (Pass)
R^2	0.92	0.92	0.92	0.92
Number of Measurements	169	169	167	167
Root Mean Square Error (µg m^{-3})	3.1	1.9	3.0	1.7
Mean Absolute Error (µg m^{-3})	2.7	1.4	2.6	1.3

Figure 13 Orthogonal regression plot – all uncorrected FIDAS (Method 11) PM_{10} data

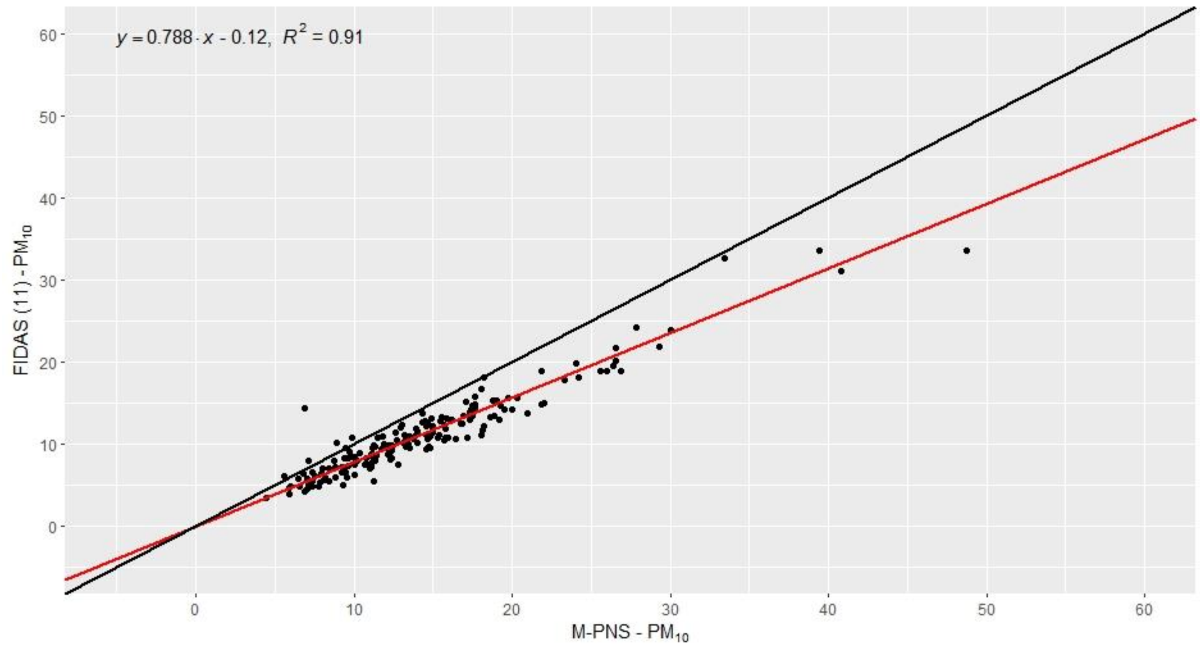


Figure 14 Orthogonal regression plot – uncorrected FIDAS (Method 11) PM_{10} data with two outliers removed

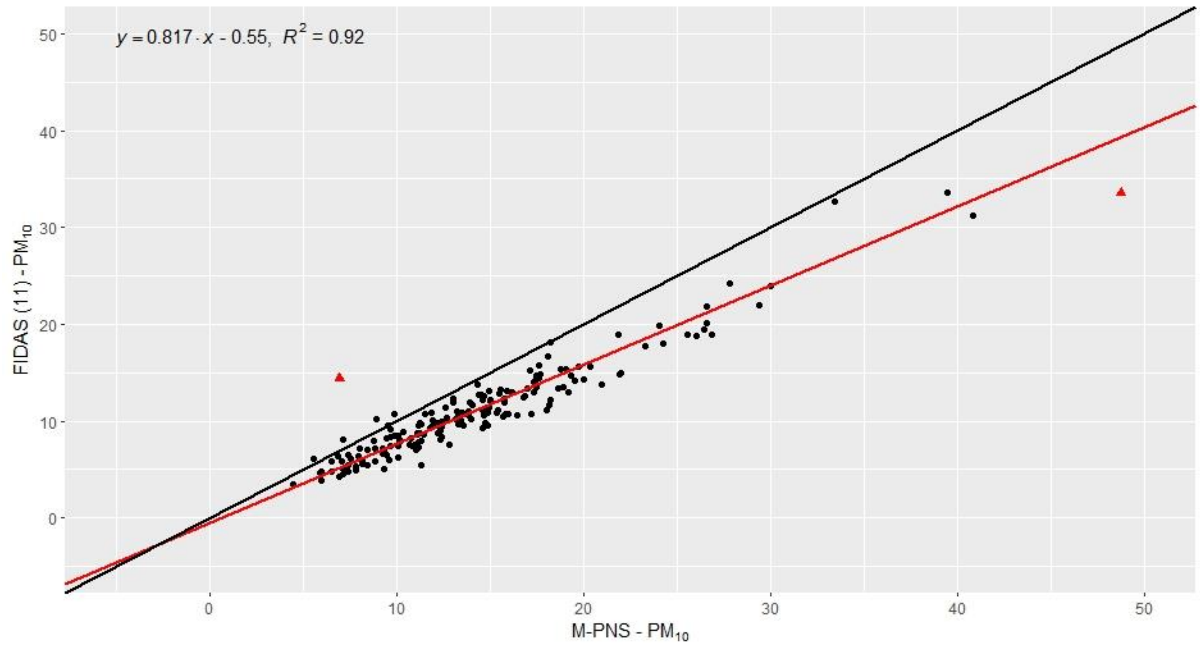


Figure 15 Orthogonal regression plot – FIDAS (Method 11) PM_{10} divided by 0.817

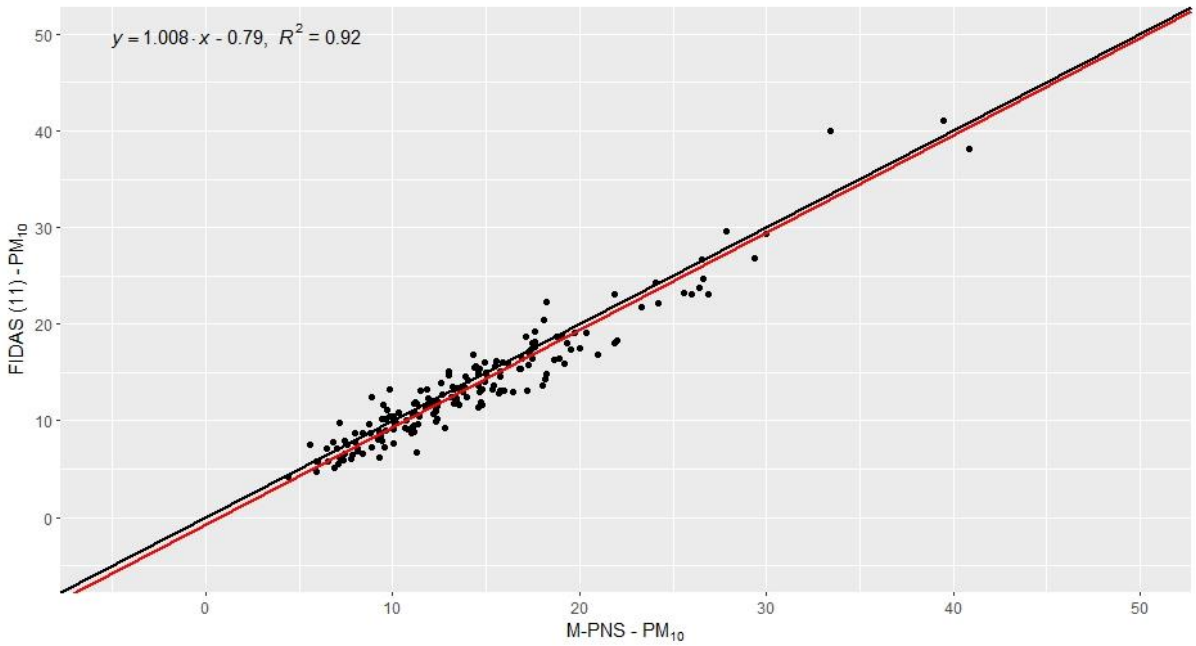


Table 21 Summary results – Fidas (Method 11) PM_{10}

Measure	Uncorrected Data	Corrected Data (Divided by 0.788)	Uncorrected Data - Outliers Removed	Corrected Data - Outliers Removed (Divided by 0.817)
Intercept	-0.119 ± 0.283 (Not Significant)	-0.314 ± 0.36 (Not Significant)	-0.549 ± 0.268 (Significant)	-0.789 ± 0.328 (Significant)
Slope	0.788 ± 0.018 (Significant)	1.011 ± 0.023 (Not Significant)	0.817 ± 0.017 (Significant)	1.008 ± 0.021 (Not Significant)
Expanded Uncertainty at 50 µg m^{-3} (%)	43.2 (Fail)	8.5 (Pass)	39.1 (Fail)	7.4 (Pass)
R^2	0.91	0.91	0.92	0.92
Number of Measurements	178	178	176	176
Root Mean Square Error (µg m^{-3})	3.9	2.0	3.7	1.7
Mean Absolute Error (µg m^{-3})	3.3	1.4	3.2	1.3

Figure 16 Orthogonal regression plot – all uncorrected FIDAS (Method 73) PM_{10} data

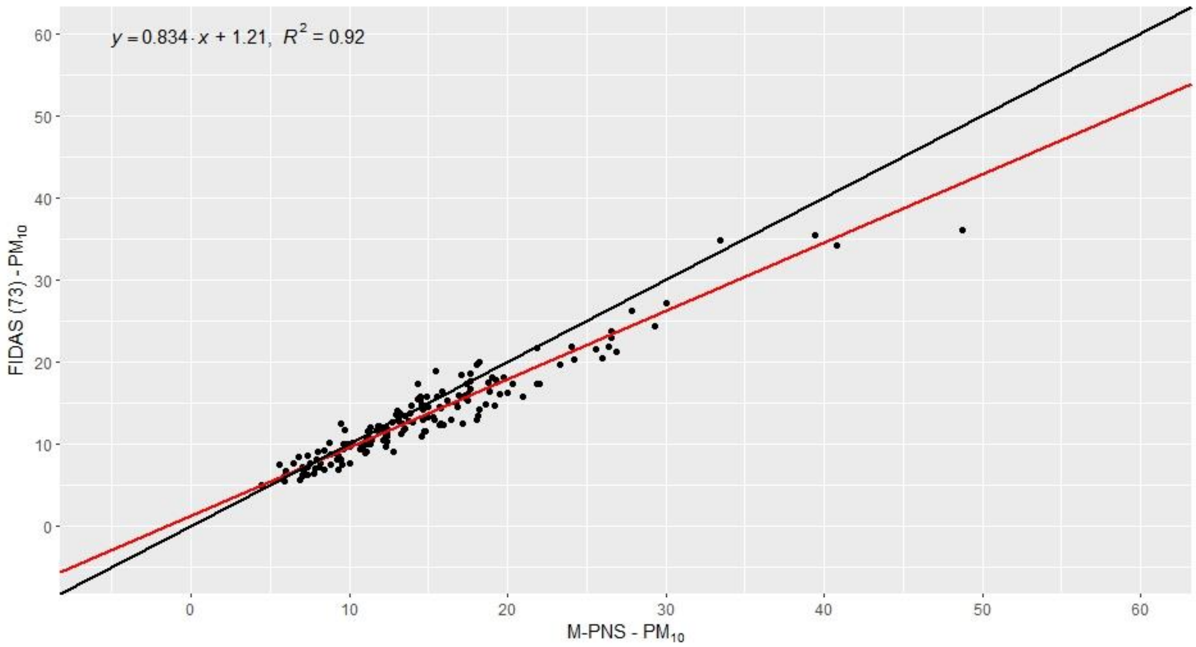


Figure 17 Orthogonal regression plot – uncorrected FIDAS (Method 73) PM_{10} data with one outlier removed

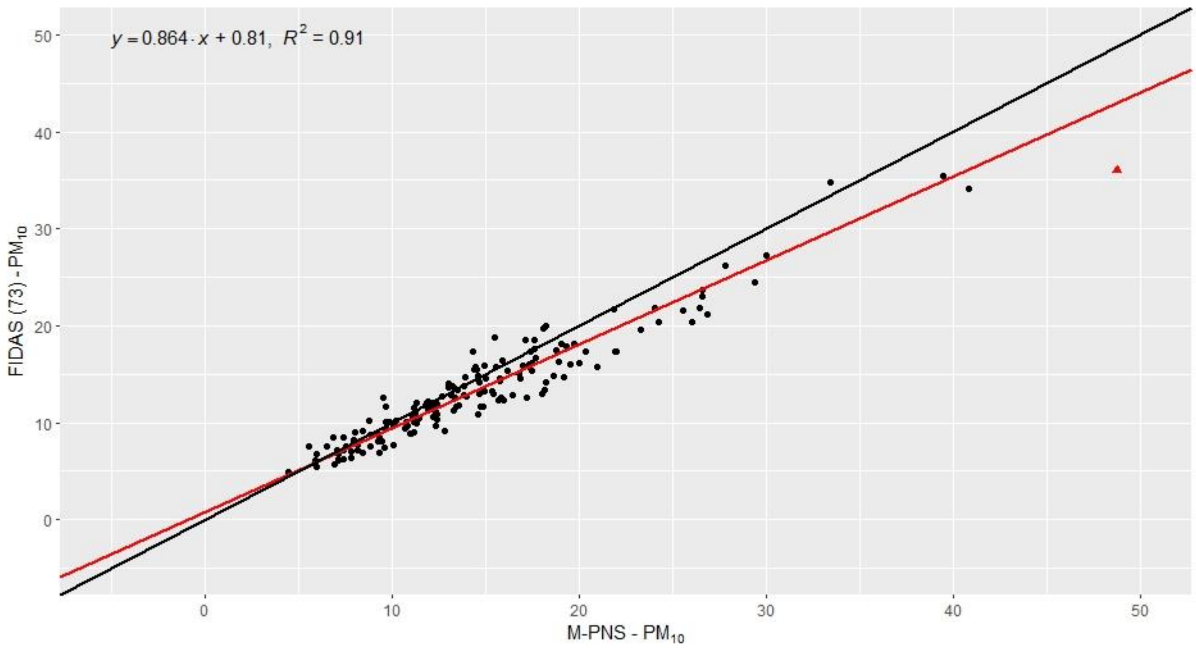


Figure 18 Orthogonal regression plot – FIDAS (Method 73) PM_{10} divided by 0.864

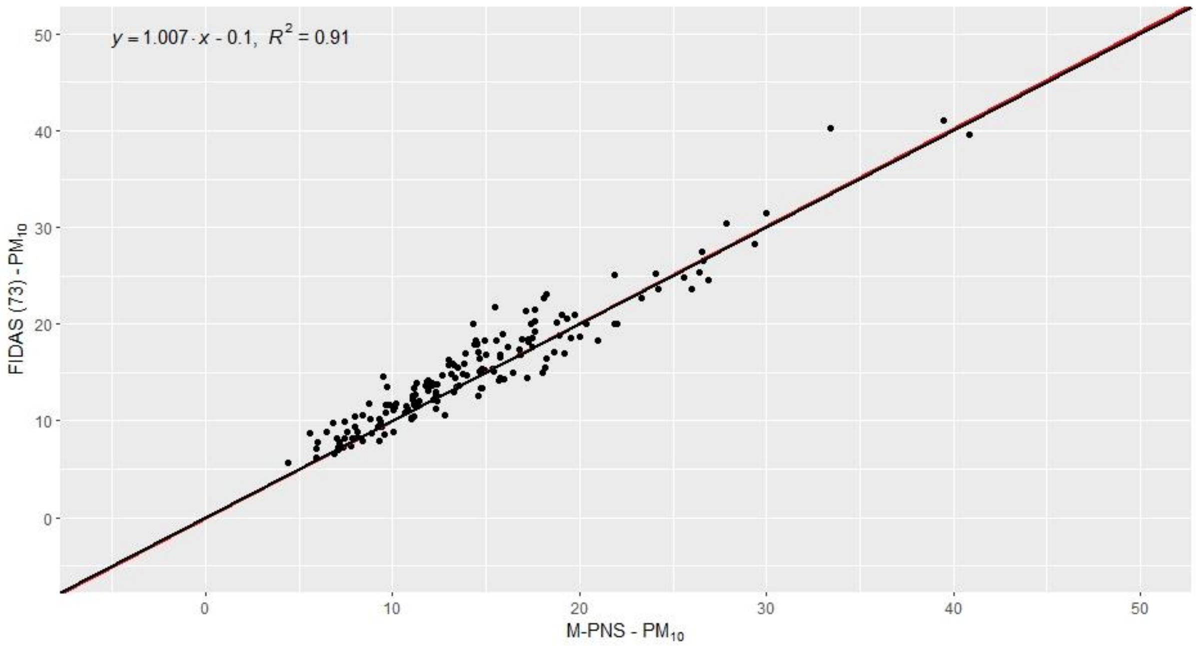


Table 22 Summary results – Fidas (Method 73) PM_{10}

Measure	Uncorrected Data	Corrected Data (Divided by 0.834)	Uncorrected Data - Outliers Removed	Corrected Data - Outliers Removed (Divided by 0.864)
Intercept	1.212 ± 0.308 (Significant)	1.337 ± 0.369	0.805 ± 0.317 (Significant)	0.836 ± 0.367
Slope	0.834 ± 0.019 (Significant)	1.008 ± 0.023	0.864 ± 0.02 (Significant)	1.007 ± 0.023
Expanded Uncertainty at 50 µg m^{-3} (%)	29.025 (Fail)	10.851 (Pass)	24.586 (Pass)	9.24 (Pass)
R^2	0.92	0.92	0.92	0.92
Number of Measurements	161	161	160	160
Root Mean Square Error (µg m^{-3})	2.4	2.0	2.2	1.8
Mean Absolute Error (µg m^{-3})	1.8	1.5	1.7	1.4

A2 $PM_{2.5}$ Regression Plots

Figures 18 to 28 show the $PM_{2.5}$ orthogonal regression plots for the comparison between the MPNS reference sampler and the four automatic methods. The automatic data have been corrected based on the significance results detailed in Table 3. Plots of both uncorrected data and corrected data are shown – in this case BAM $PM_{2.5}$ data did not require any correction and so only uncorrected data are shown (Figure 19).

Two lines are shown in the plots:

- The black 1:1 line.
- The red orthogonal regression line.

For the plots with the outliers removed, the identified outliers are shown as red triangles – see Figure 20 as an example. A summary of the regression analysis is shown in Tables 22 – 25.

Figure 19 Orthogonal regression plot – uncorrected BAM $PM_{2.5}$ data

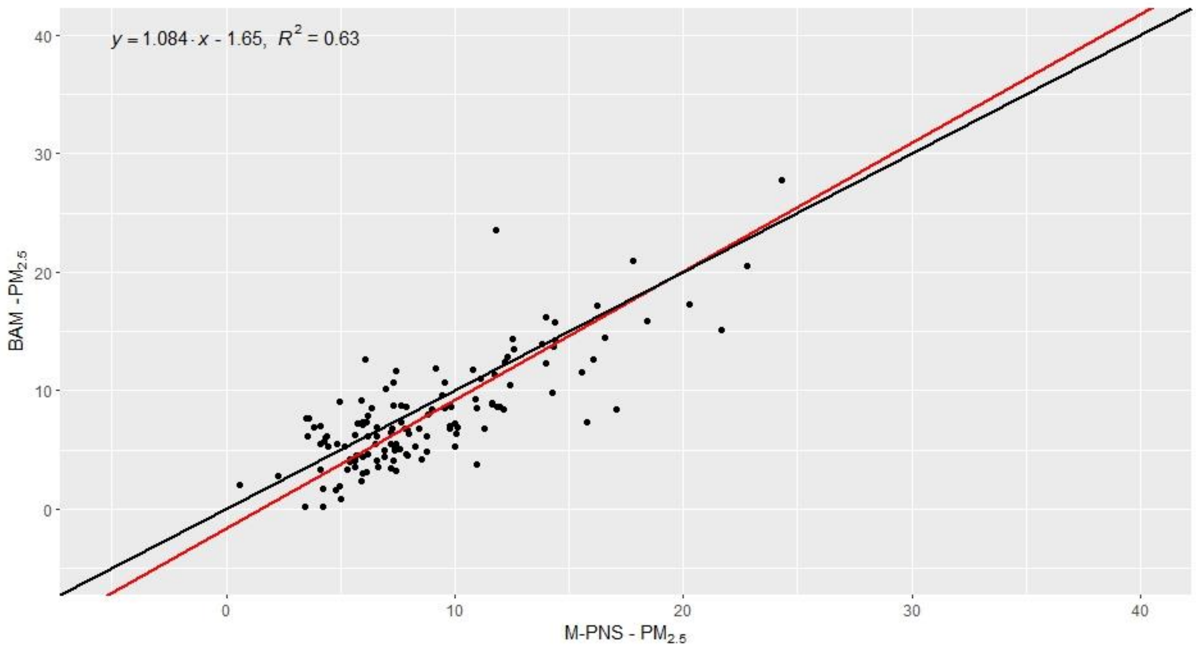


Figure 20 Orthogonal regression plot – uncorrected BAM $PM_{2.5}$ data with one outlier removed

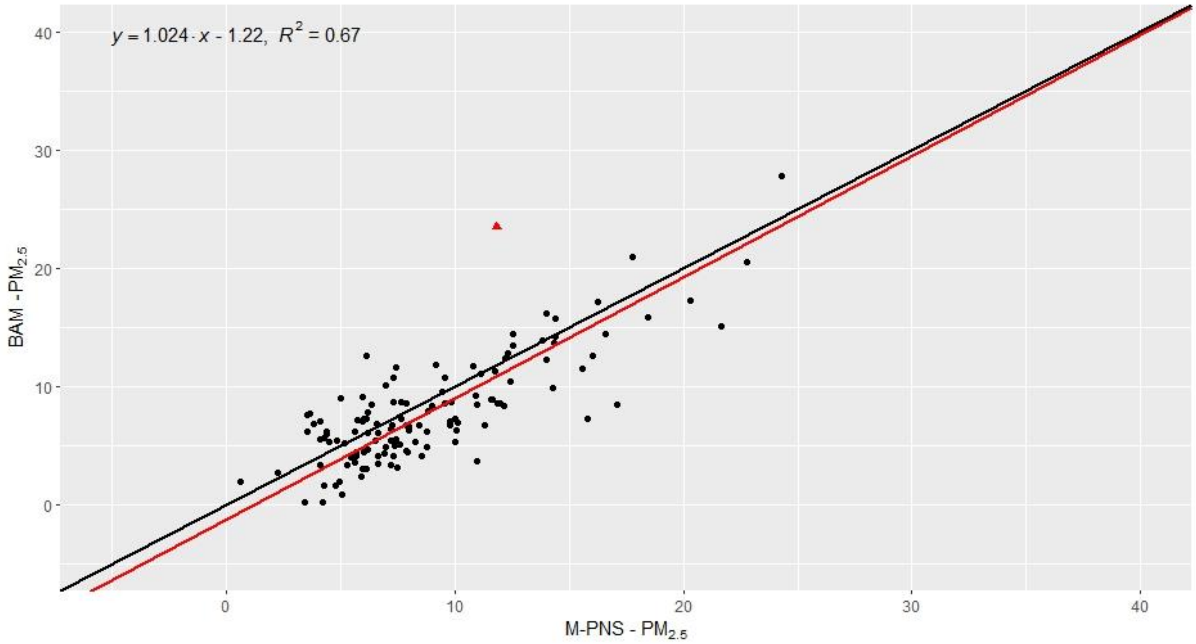


Table 23 Summary results – BAM $PM_{2.5}$

Measure	Uncorrected Data	Uncorrected Data - Outliers Removed
Intercept	-1.653 ± 0.57 (Significant)	-1.216 ± 0.52 (Significant)
Slope	1.084 ± 0.058 (Not Significant)	1.024 ± 0.053 (Not Significant)
Expanded Uncertainty at 30 µg m^{-3} (%)	20.4 (Pass)	17.8 (Pass)
R^2	0.63	0.67
Number of Measurements	127	126
Root Mean Square Error (µg m^{-3})	3.0	2.8
Mean Absolute Error (µg m^{-3})	2.4	2.3

Figure 21 Orthogonal regression plot – uncorrected BAM $PM_{2.5}$ data with four outliers removed

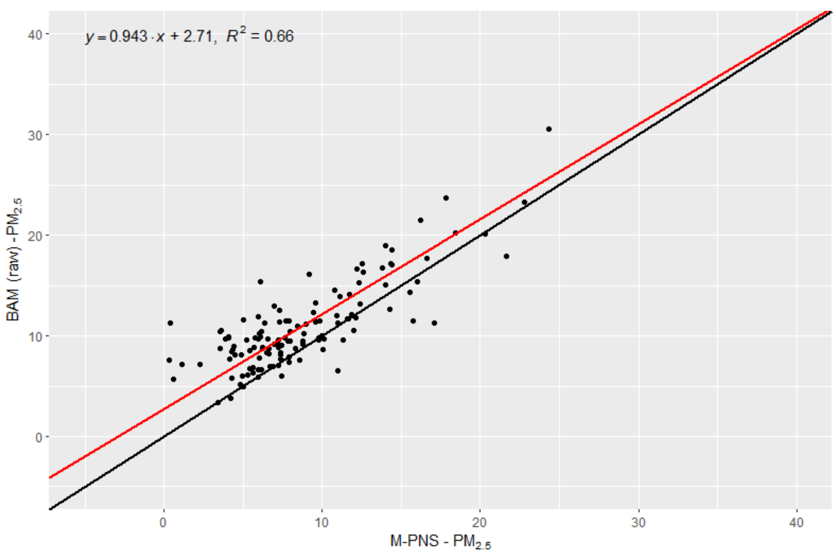


Figure 22 Orthogonal regression plot – all uncorrected FDMS $PM_{2.5}$ data

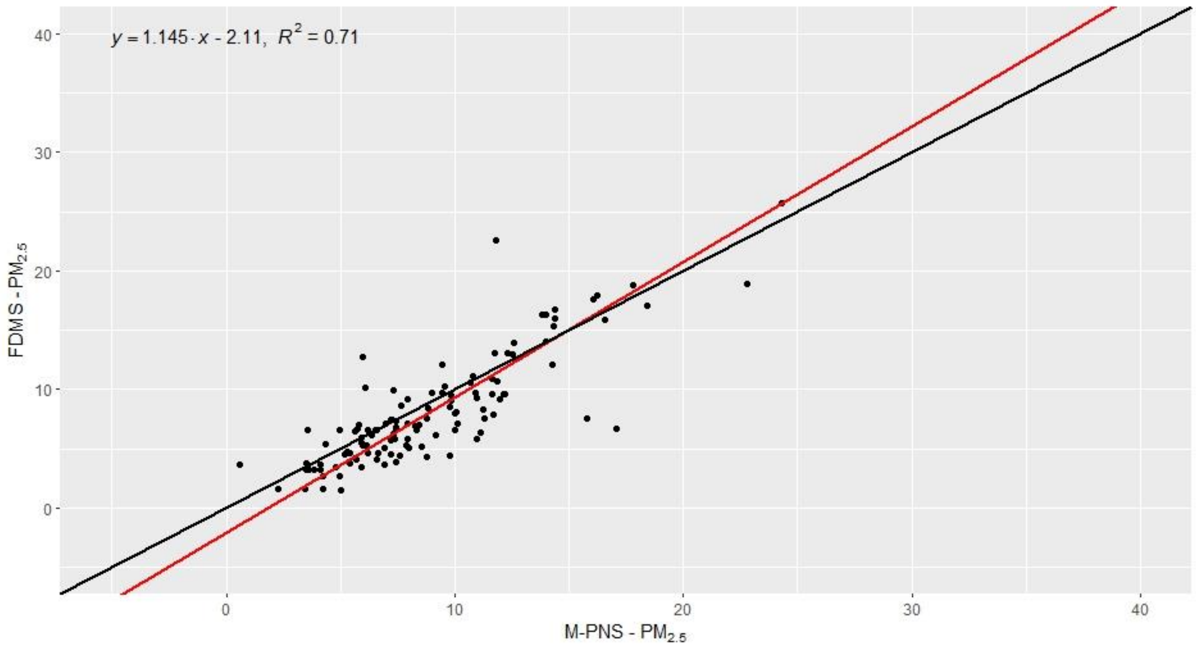


Figure 23 Orthogonal regression plot – uncorrected FDMS $PM_{2.5}$ data with four outliers removed

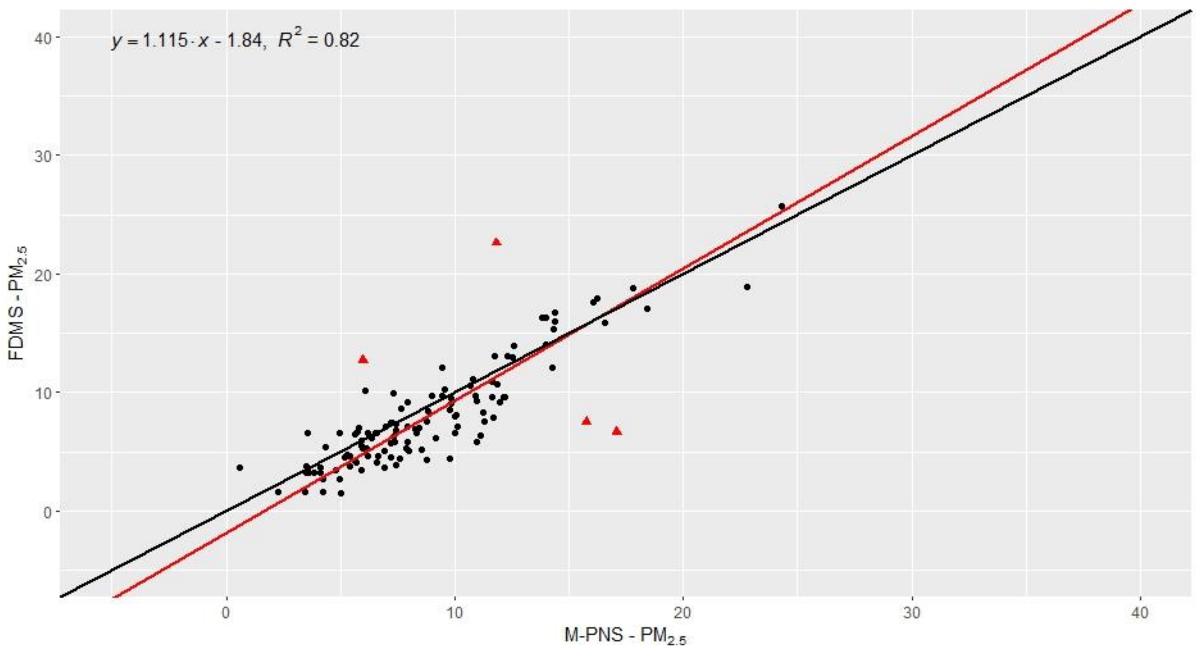


Figure 24 Orthogonal regression plot – FDMS $PM_{2.5}$ divided by 1.115

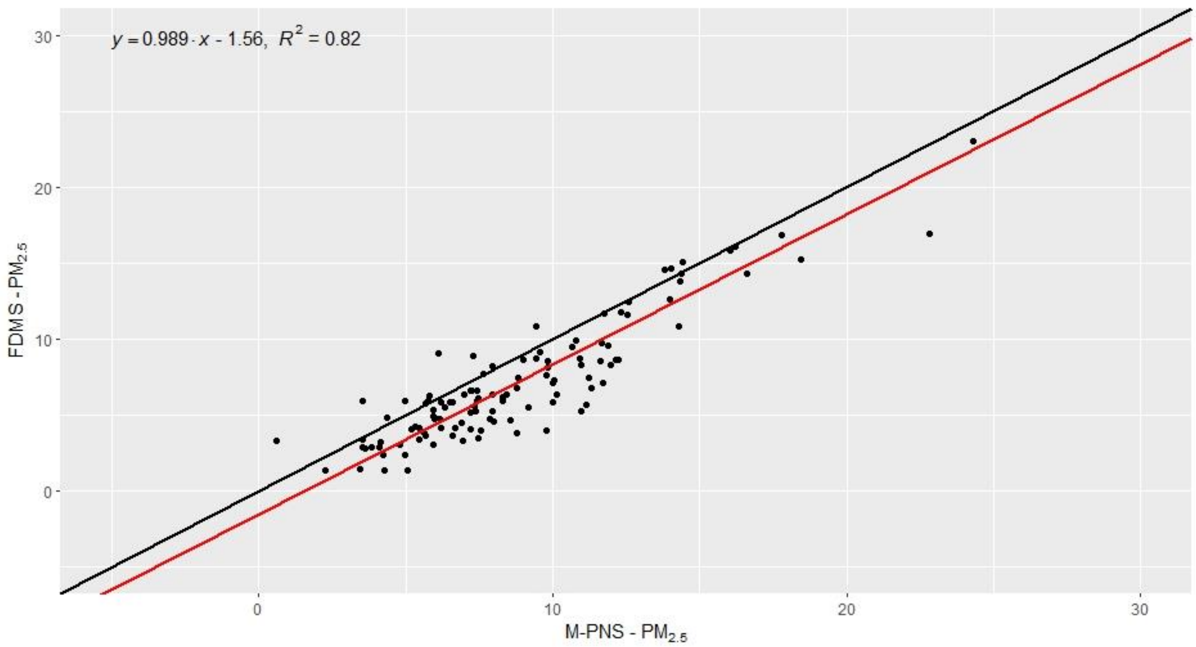


Table 24 Summary results – FDMS $PM_{2.5}$

Measure	Uncorrected Data	Corrected Data (Divided by 1.145)	Uncorrected Data - Outliers Removed	Corrected Data - Outliers Removed (Divided by 1.115)
Intercept	-2.107 ± 0.543 (Significant)	-1.618 ± 0.474	-1.844 ± 0.415 (Significant)	-1.559 ± 0.372
Slope	1.145 ± 0.056 (Significant)	0.975 ± 0.049	1.115 ± 0.043 (Significant)	0.989 ± 0.039
Expanded Uncertainty at 30 µg m^{-3} (%)	22.5 (Pass)	24.1 (Pass)	16.0 (Pass)	18.6 (Pass)
R^2	0.71	0.71	0.82	0.82
Number of Measurements	120	120	116	116
Root Mean Square Error (µg m^{-3})	2.6	2.3	2.0	1.7
Mean Absolute Error (µg m^{-3})	1.9	1.6	1.6	1.3

Figure 25 Orthogonal regression plot – FIDAS (Method 11) $PM_{2.5}$ (uncorrected)

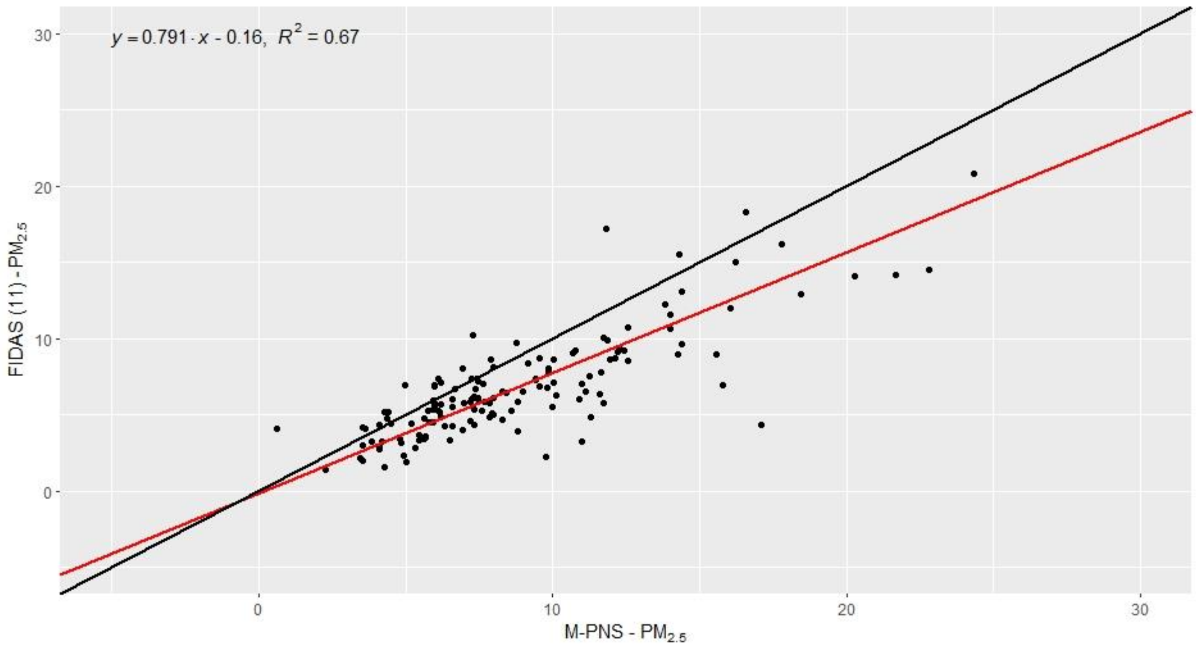


Figure 26 Orthogonal regression plot – FIDAS (Method 11) $PM_{2.5}$ with one outlier removed

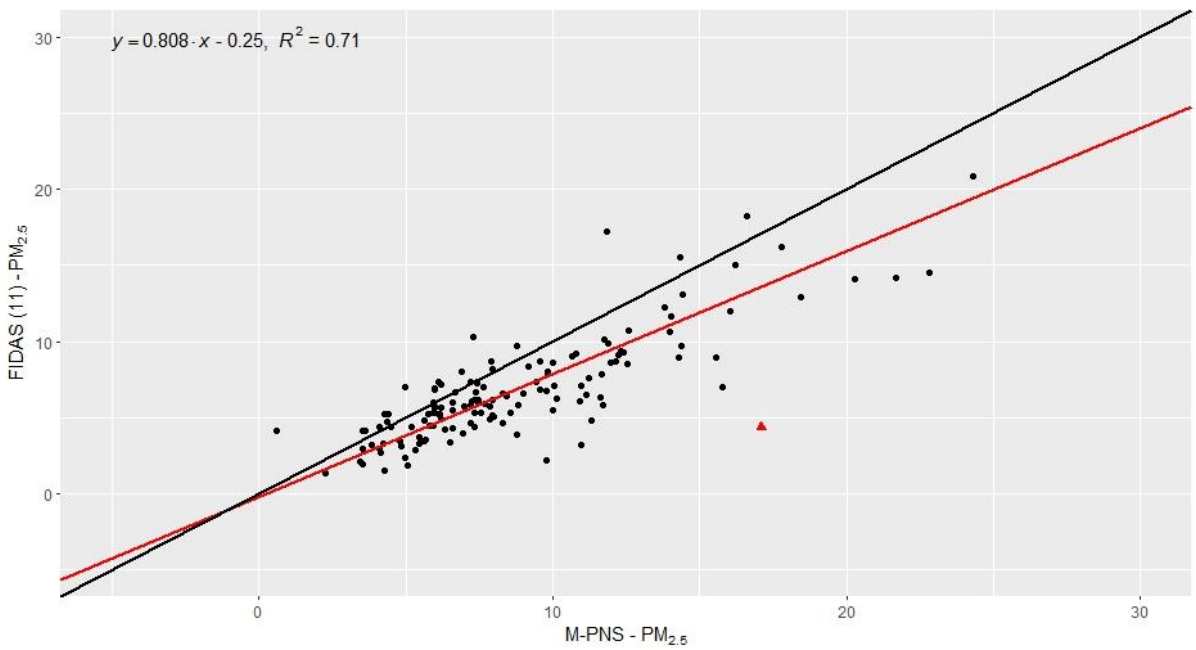


Figure 27 Orthogonal regression plot – FIDAS (Method 11) $PM_{2.5}$ divided by 0.808

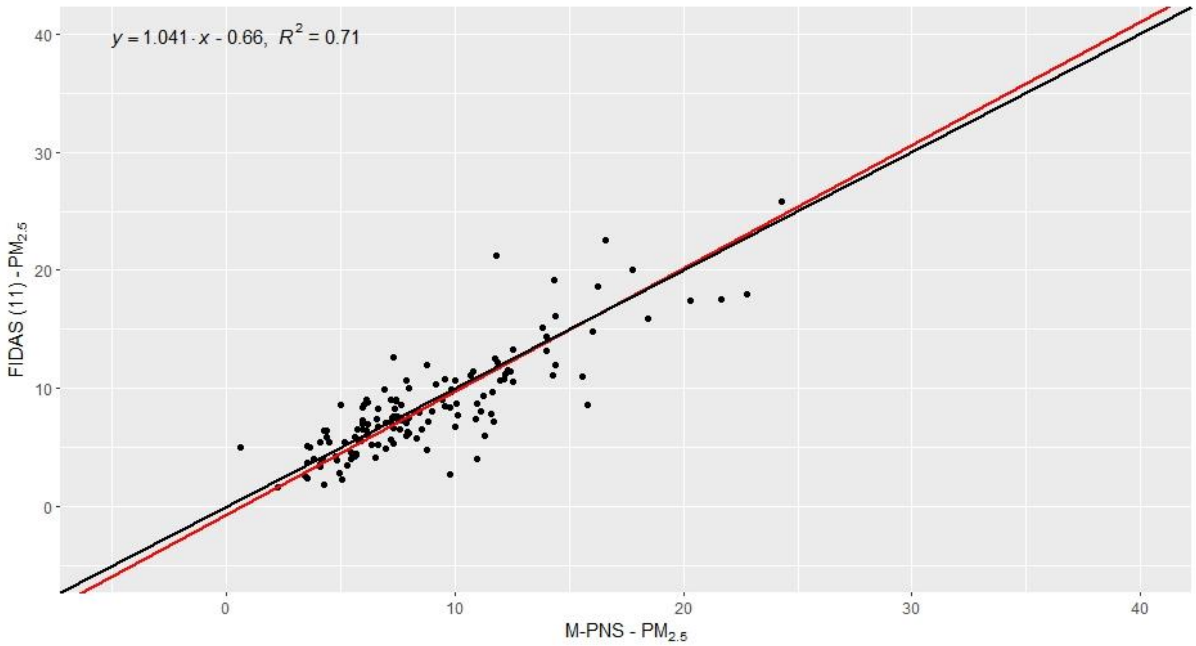


Table 25 Summary results – Fidas (Method 11) $PM_{2.5}$

Measure	Uncorrected Data	Corrected Data (Divided by 0.791)	Uncorrected Data - Outliers Removed	Corrected Data - Outliers Removed (Divided by 0.808)
Intercept	-0.163 ± 0.398 (Not Significant)	-0.672 ± 0.503 (Not Significant)	-0.245 ± 0.377 (Not Significant)	-0.658 ± 0.466 (Not Significant)
Slope	0.791 ± 0.041 (Significant)	1.053 ± 0.051 (Not Significant)	0.808 ± 0.039 (Significant)	1.041 ± 0.048 (Not Significant)
Expanded Uncertainty at 30 µg m^{-3} (%)	44.8 (Fail)	20.1 (Pass)	41.9 (Fail)	17.9 (Pass)
R^2	0.67	0.67	0.71	0.71
Number of Measurements	138	138	137	137
Root Mean Square Error (µg m^{-3})	3.2	2.6	3.0	2.4
Mean Absolute Error (µg m^{-3})	2.4	1.9	2.3	1.8

Figure 28 Orthogonal regression plot – FIDAS (Method 73) $PM_{2.5}$ (Uncorrected)

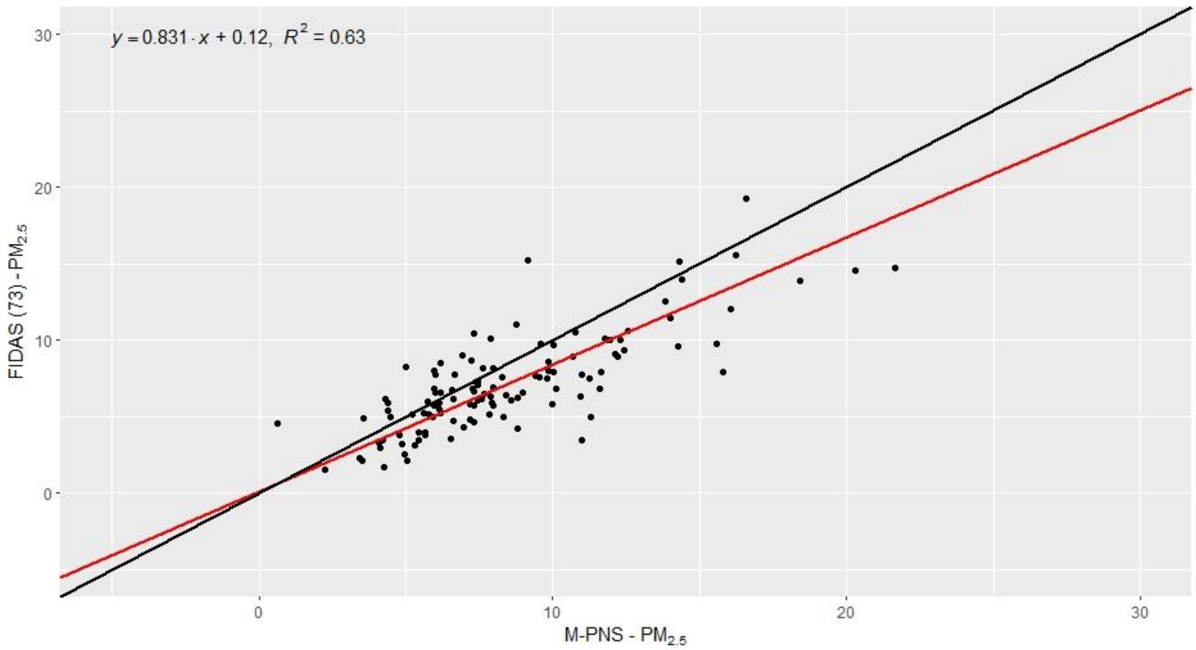


Figure 29 Orthogonal regression plot – uncorrected FIDAS (Method 73) $PM_{2.5}$ with one outlier removed

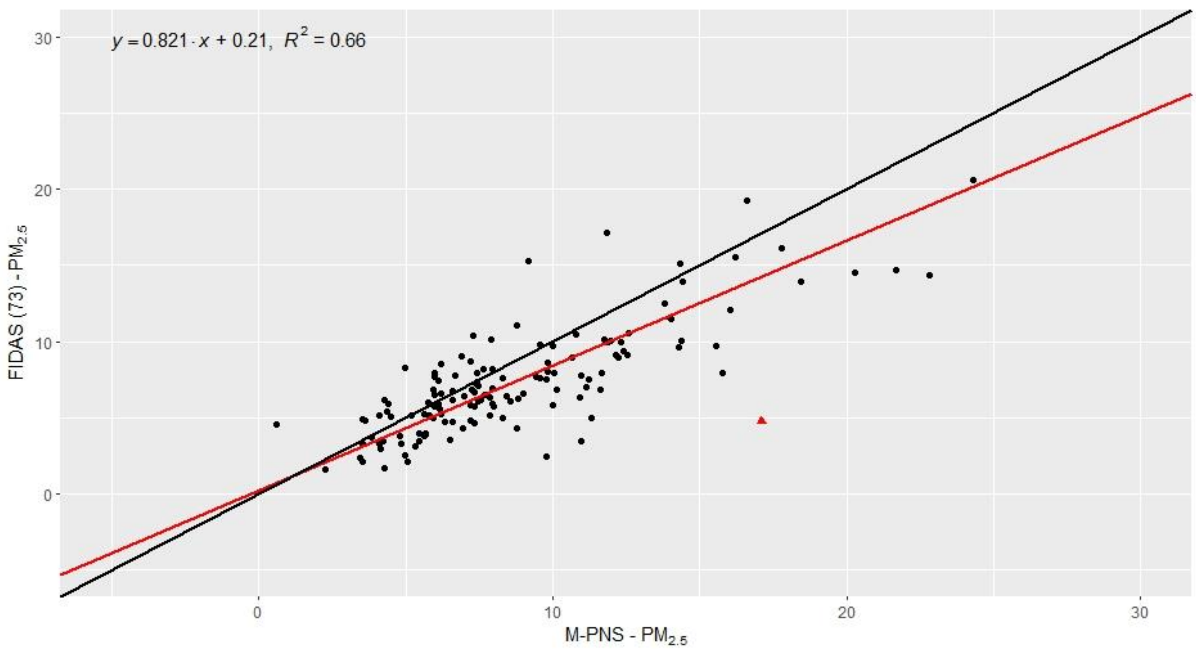


Figure 30 Orthogonal regression plot – FIDAS (Method 73) $PM_{2.5}$ divided by 0.821

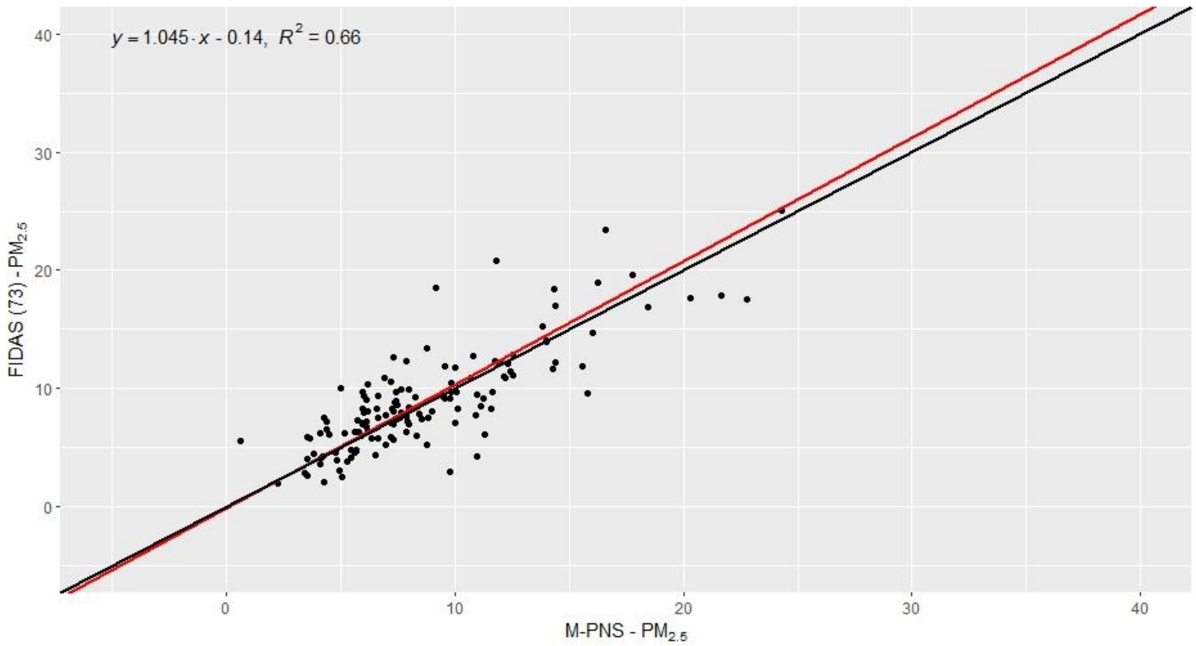


Table 26 Summary results – Fidas (Method 73) $PM_{2.5}$

Measure	Uncorrected Data	Corrected Data (Divided by 0.791)	Uncorrected Data - Outliers Removed	Corrected Data - Outliers Removed (Divided by 0.808)
Intercept	0.293 ± 0.434 (Not Significant)	-0.151 ± 0.539	0.21 ± 0.414 (Not Significant)	-0.14 ± 0.504
Slope	0.804 ± 0.044 (Significant)	1.059 ± 0.055	0.821 ± 0.043 (Significant)	1.045 ± 0.052
Expanded Uncertainty at 30 µg m-3 (%)	40.0 (Fail)	23.4 (Pass)	37.0 (Fail)	20.9 (Pass)
R2	0.63	0.63	0.66	0.66
Number of Measurements	136	136	135	135
Root Mean Square Error (µg m-3)	3.0	2.8	2.8	2.6
Mean Absolute Error (µg m-3)	2.2	2.0	2.2	1.9

A3 Measurement Uncertainty – Corrected Data

Figures 29 to 36 show the expanded relative measurement uncertainty as a function of daily mean PM_{10} and $PM_{2.5}$ concentrations. The red line shows the 25% line at which measurements become equivalent. This is important in demonstrating that the measurement uncertainty is not constant with measured concentration at low concentrations. As the measured concentration decreases the measurement uncertainty begins to increase exponentially, which has a more significant impact if we are measuring PM_{10} and $PM_{2.5}$ concentrations below the Scottish annual mean objectives of 18 and 10 $\mu g\ m^{-3}$, respectively. The performance of the reference method will also play a considerable role in the total uncertainty at low concentrations.

Figure 31 Expanded uncertainty plot – BAM PM_{10} (uncorrected)

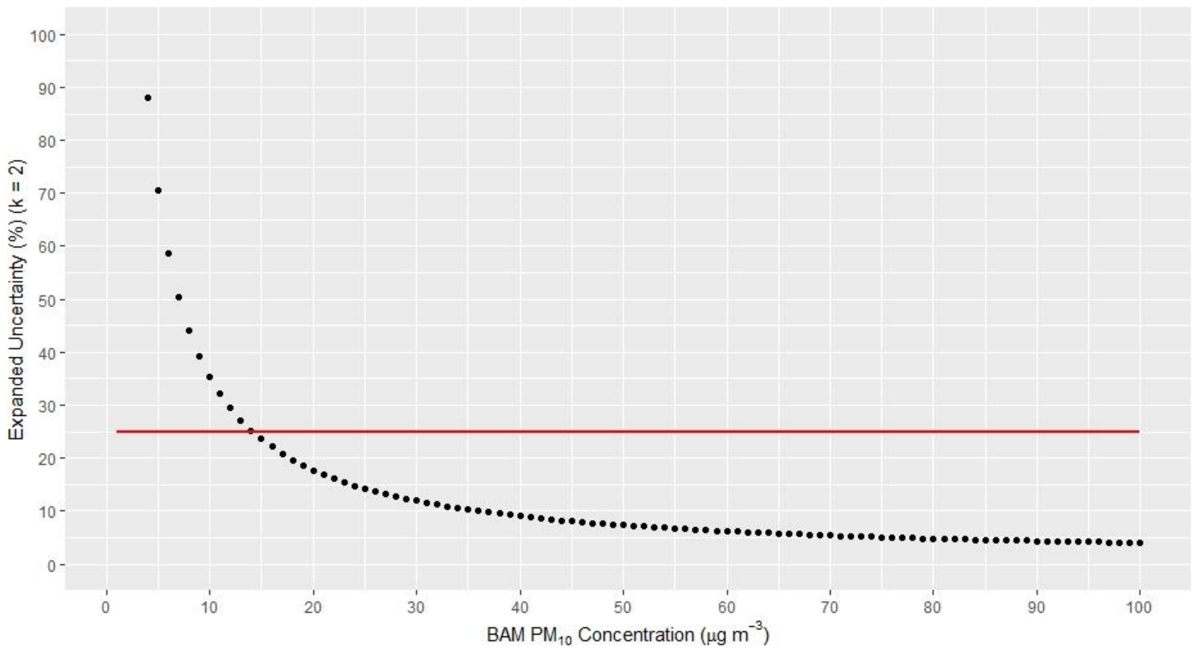


Figure 32 Expanded uncertainty plot – FDMS PM_{10} divided by 0.888

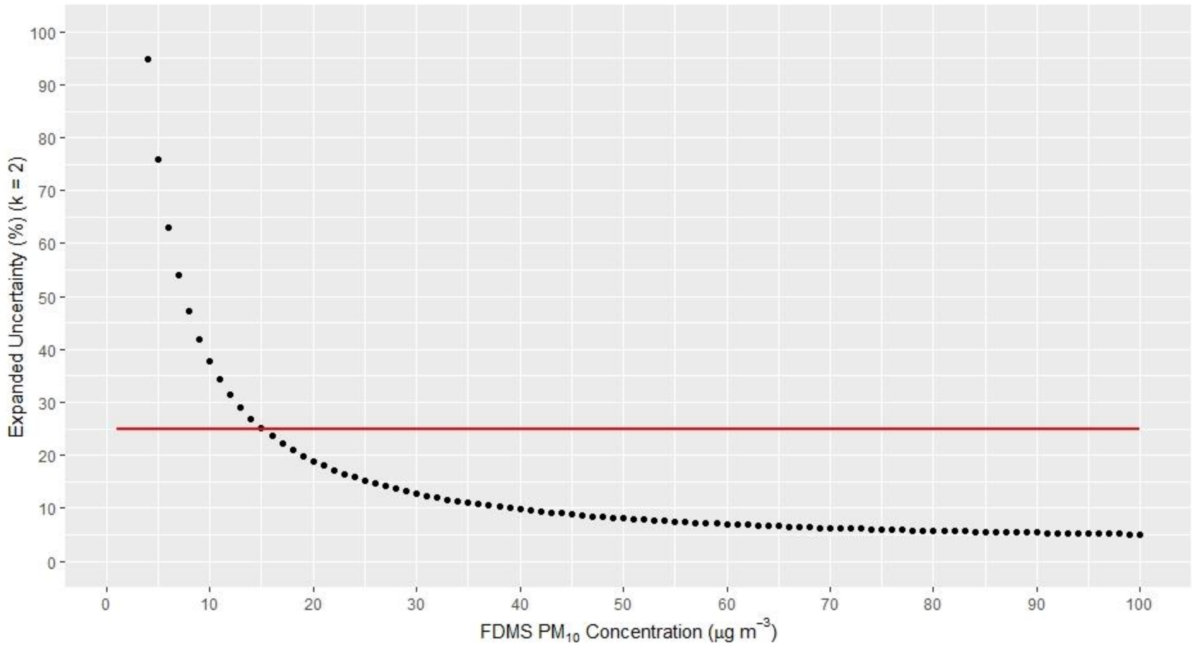


Figure 33 Expanded uncertainty plot – Fidas (Method 11) PM_{10} divided by 0.817

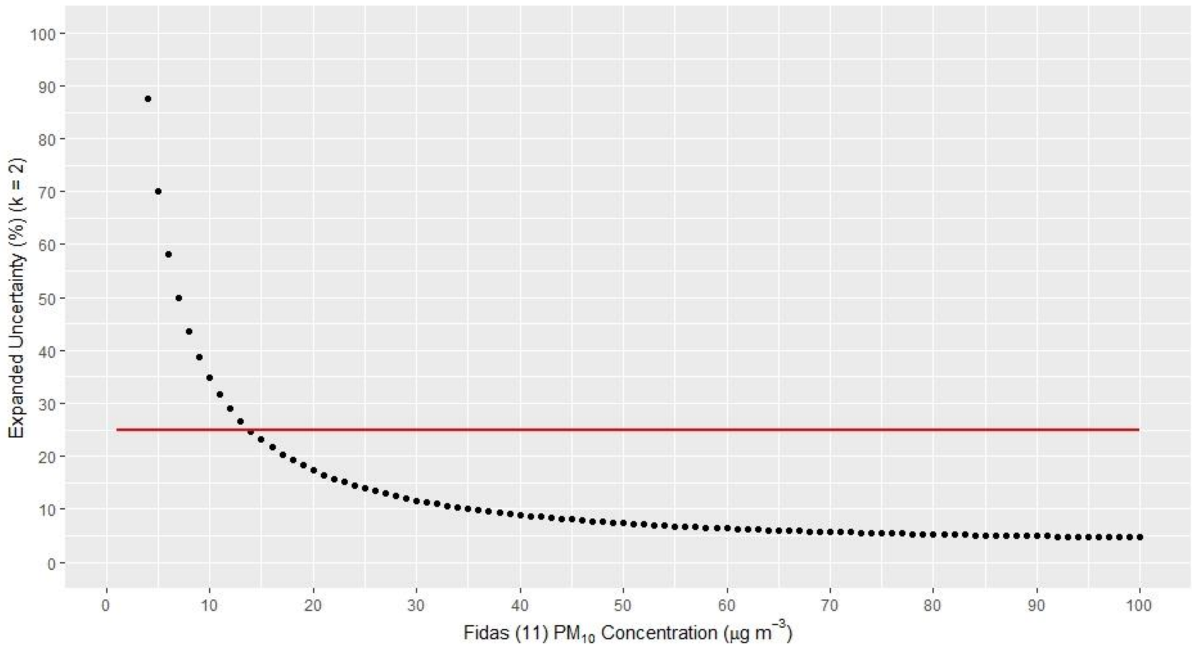


Figure 34 Expanded uncertainty plot – Fidas (Method 73) PM_{10} divided by 0.864

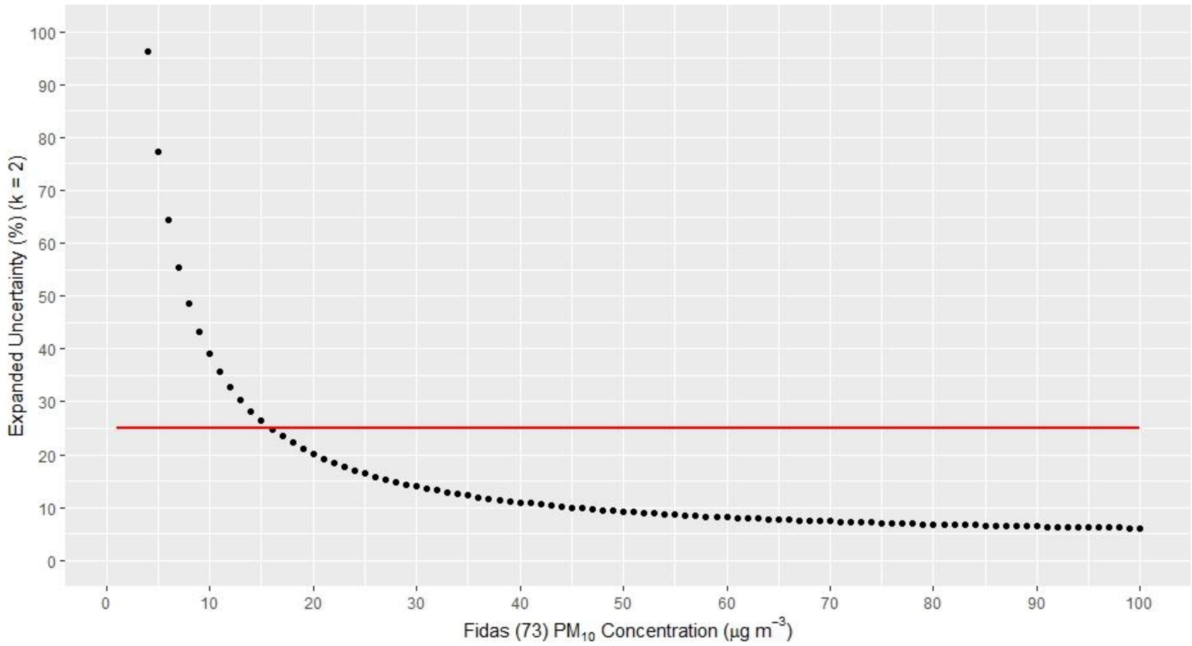


Figure 35 Expanded uncertainty plot – BAM $PM_{2.5}$ (uncorrected)

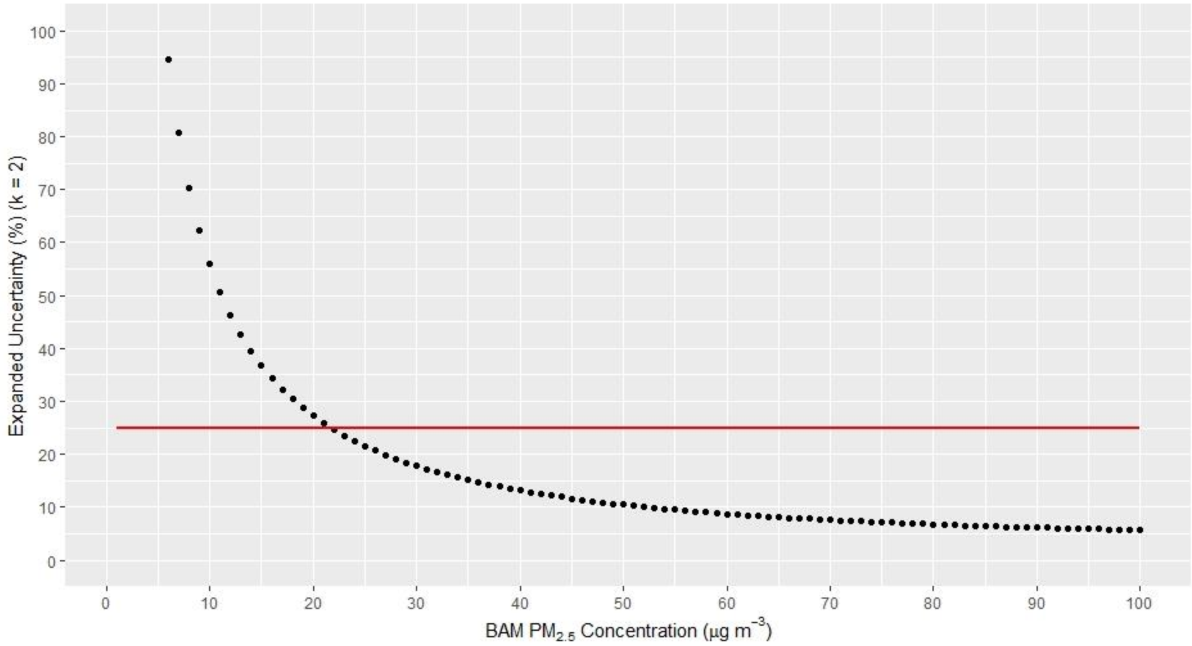


Figure 36 Expanded uncertainty plot – FDMS $PM_{2.5}$ (uncorrected)

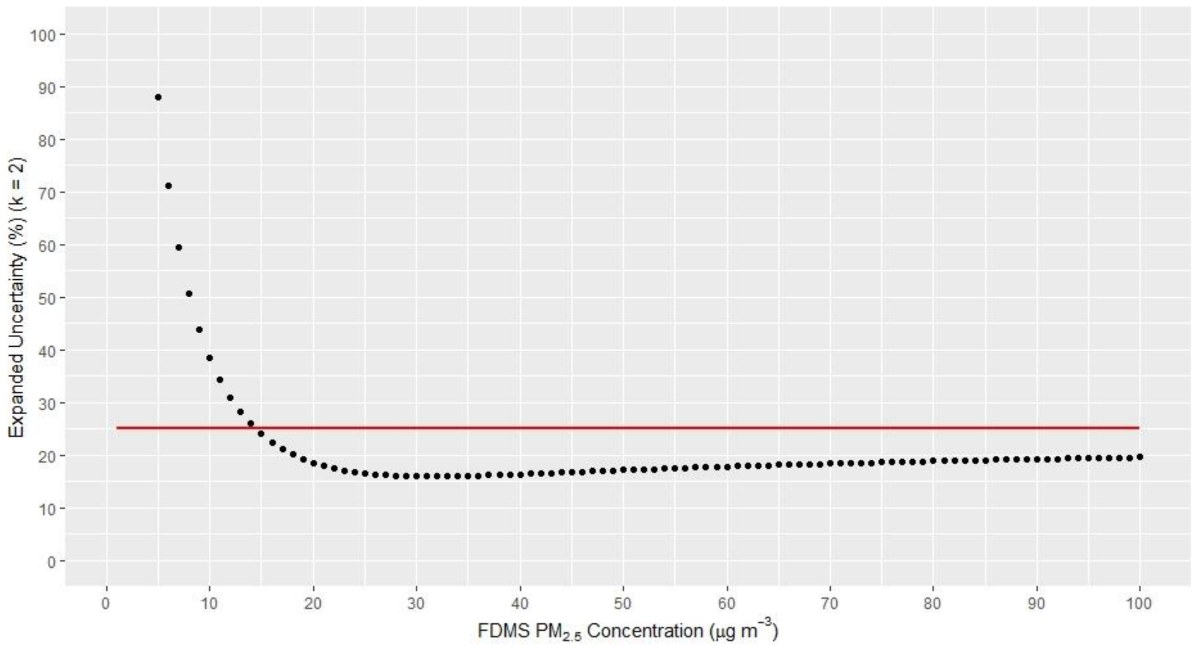


Figure 37 Expanded uncertainty plot – Fidas (Method 11) $PM_{2.5}$ divided by 0.808

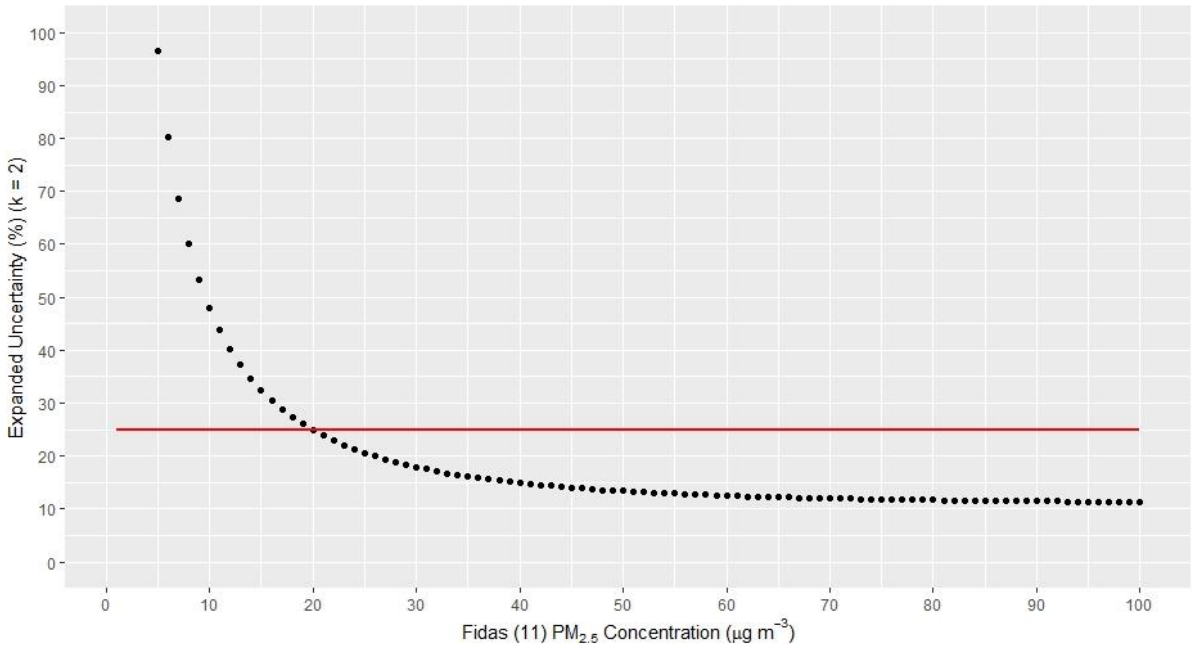
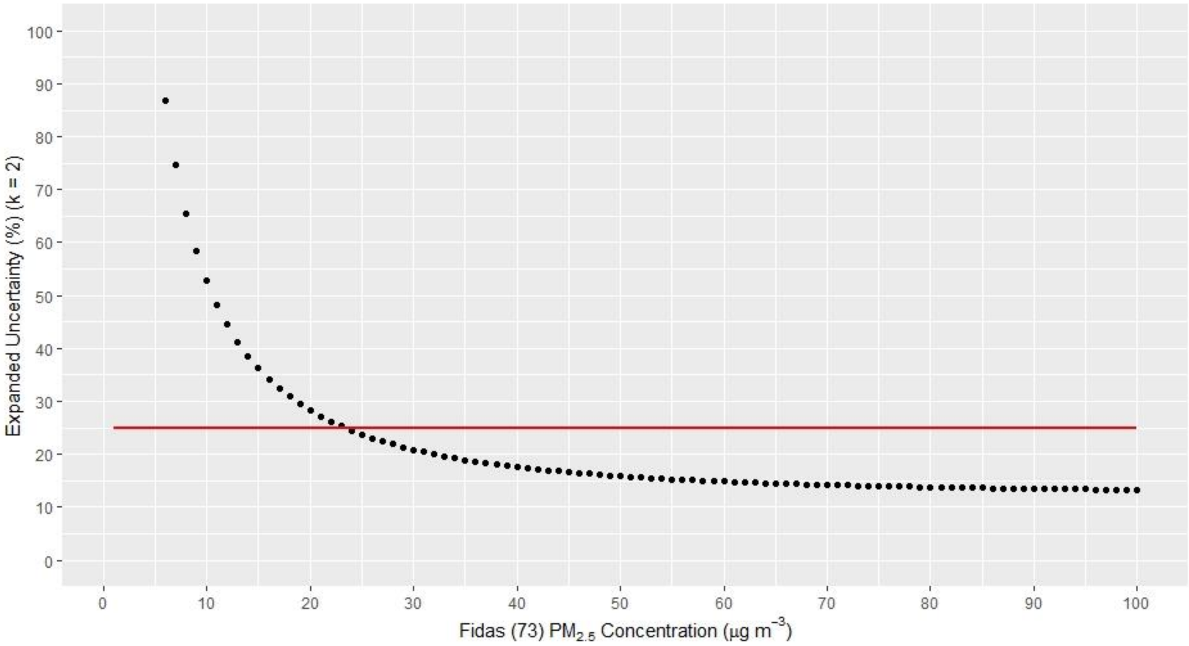


Figure 38 Expanded uncertainty plot – Fidas (Method 73) $PM_{2.5}$ divided by 0.821



A4 Calculations used for regression analysis and measurement uncertainty analysis

A4.1 Orthogonal Regression

The response of all samplers used within this study is assumed to be linear and therefore follow the equation of a straight line:

$$y_i = a + bx_i \qquad \text{(A1)}$$

Where:

x_i = concentration of the MPNS reference sampler measured at point i, where i > 0.

y_i = concentration of the automatic analyser measure at point i, where i > 0.

a = intercept.

b = slope.

The following equations were used to calculate the slope and intercept for the best-fit orthogonal regression line:

$$b = \frac{S_{yy} - S_{xx} + \left[\left(S_{yy} - S_{xx}\right)^2 + 4\left(S_{xy}\right)^2\right]^{\frac{1}{2}}}{2S_{xy}} \qquad \text{(A2)}$$

$$a = \bar{y} - b \cdot \bar{x} \qquad \text{(A3)}$$

Where:

$$S_{xx} = \sum_{i=1}^{i=n} (x_i - \bar{x})^2 \qquad \text{(A4)}$$

$$S_{yy} = \sum_{i=1}^{i=n} (y_i - \bar{y})^2 \qquad \text{(A5)}$$

$$S_{xy} = \sum_{i=1}^{i=n} (x_i - \bar{x}) \cdot (y_i - \bar{y}) \qquad \text{(A6)}$$

Where n is the number of paired measurements recorded.

$$\bar{x} = \frac{\sum_{i=1}^{i=n} x_i}{n} \qquad \text{(A7)}$$

$$\bar{y} = \frac{\sum_{i=1}^{i=n} y_i}{n} \qquad \text{(A8)}$$

The coefficient of determination, r^2, is calculated using the following equation:

$$r^2 = \left[\frac{\left[n \sum_{i=1}^{i=n}(xy)\right] - \left[\sum_{i=1}^{i=n} x \cdot \sum_{i=1}^{i=n} y\right]}{\left[\left(n \sum_{i=1}^{i=n}(x^2)\right) - \left(\sum_{i=1}^{i=n} x\right)^2\right] \cdot \left[\left(n \sum_{i=1}^{i=n}(y^2)\right) - \left(\sum_{i=1}^{i=n} y\right)^2\right]}\right]^2 \qquad \text{(A9)}$$

A4.2 Uncertainty in Slope and Intercept – Uncorrected Datasets

The uncertainty in the slope (u_b) is calculated using:

$$u_b = \left[\frac{S_{yy} - ({S_{xy}}^2/S_{xx}}{(n-2)S_{xx}}\right]^{\frac{1}{2}} \quad \text{(A10)}$$

The uncertainty in the intercept (u_a) is calculated using:

$$u_a = \left[{u_b}^2 \frac{\sum_{i=1}^{i=n} {x_i}^2}{n}\right]^{\frac{1}{2}} \quad \text{(A11)}$$

A4.3 Significance of Intercept and Slope

In order to determine if data need to be corrected for intercept (a) or slope (b) using the colocation results, the following criteria have been used to define if these are significant. The calculate intercepts and slopes are ***not*** deemed significant if:

$$|a| \leq 2u_a \quad \text{(A12)}$$

$$|b-1| \leq 2u_b \quad \text{(A13)}$$

In this case no correction for intercept or slope is made.

A4.4 Uncertainty in Measurements

The residual sum of squares (RSS) from the orthogonal regression is calculated using:

$$RSS = \sum_{i=1}^{i=n}(y_i - a - bx_i)^2 \quad \text{(A14)}$$

The uncertainty in each y-value $(\text{Random Term}, \sigma)$ is calculated using:

$$\sigma = \left[\frac{1}{n-2}\sum_{i=1}^{i=n}(y_i - a - bx_i)^2 - {u_{RM}}^2\right]^{\frac{1}{2}} \quad \text{(A15)}$$

Where u_{RM} is the random uncertainty of the reference method and defaults to 0.67 μg m^{-3} for this study as only one reference sampler was used.

The uncertainty in the results $(u(y_i))$ is calculated using:

$$[u(y_i)]^2 = \sigma^2 + [a + (b-1)z_i]^2 \quad \text{(A16)}$$

Where z_i is the reference concentration at which the uncertainty is calculated and $a + (b-1)z_i$ is the Bias (B_i).

The combined relative uncertainty of the results $(w_h(y_i))$ is calculated using:

$$w_h(y_i) = 100 \cdot \left(\frac{u(y_i)}{z_i}\right) \quad \text{(A17)}$$

The expanded uncertainty is calculated using a coverage factor of $k = 2$ reflecting a 95% confidence interval with a normal distribution associated with the large number of measurements. Therefore:

$$W(y_i) = k \cdot w_h(y_i) = 2w_h(y_i) \quad \text{(A18)}$$

A4.5 Uncertainty in Intercept Corrected Datasets

If it is found from the colocation exercises that the data from any of the automatic analysers requires a correction for intercept then the following equation is used to calculate the corrected data $(y_{i,corr})$:

$$y_{i,corr} = y_i - a \tag{A19}$$

Orthogonal regression is then carried out using the corrected data with the equation of the straight line:

$$y_{i,corr} = c + dx_i \tag{A20}$$

Where c is the intercept calculated using the corrected data and d is the slope calculated using the corrected data.

In order to take account of the uncertainty in the intercept introduced during the colocation exercise regression (u_a) this is added to the uncertainty calculation:

$$\left[u(y_{i,corr})\right]^2 = \sigma^2 + \ [c + ((d-1)z_i]^2 + \ {u_a}^2 \tag{A21}$$

A4.6 Uncertainty in Slope Corrected Datasets

If it is found from the colocation exercises that the data from any of the automatic analysers requires require a correction for slope then the following equation is used to calculate the corrected data $(y_{i,corr})$:

$$y_{i,corr} = \frac{y_i}{b} \tag{A22}$$

In order to take account of the uncertainty in the intercept introduced during the colocation exercise regression (u_b) this is added to the uncertainty calculation:

$$\left[u(y_{i,corr})\right]^2 = \sigma^2 + \ [c + ((d-1)z_i]^2 + \ {z_i}^2{u_b}^2 \tag{A23}$$

A4.7 Uncertainty in Intercept and Slope Corrected Datasets

If it is found from the colocation exercises that the data from any of the automatic analysers require a correction for slope and intercept then the following equation is used to calculate the corrected data $(y_{i,corr})$:

$$y_{i,corr} = \frac{y_i - a}{b} \tag{A24}$$

In order to take account of the uncertainty in the intercept (u_a) and slope (u_b) introduced during the colocation exercise regression this is added to the uncertainty calculation:

$$\left[u(y_{i,corr})\right]^2 = \sigma^2 + \ [c + ((d-1)z_i]^2 + \ {z_i}^2{u_b}^2 + \ {u_a}^2 \tag{A25}$$

A4.8 Uncertainty in the Mean

The following equation is used to calculate the measurement uncertainty in the calculated mean of daily concentrations:

$$u(\bar{y}) = \left[\sum_{i=1}^{n}\left(\frac{\sigma_i}{n}\right)^2 + \sum_{i=1}^{n}(B_i)^2\right]^{1/2} \quad \text{(A28)}$$

Where:

$u(\bar{y})$ is the measurement uncertainty in the calculated mean of the automatic method (corrected)

σ_i is the calculated Random Term at the corrected candidate analyser measure at point i, where i > 0 as defined in Equation A15

B_i is the Bias at the corrected candidate analyser measure at point i, where i > 0 as defined in Equation A16

n is the number of measurements used in the calculated mean

The combined relative uncertainty of the mean $(w_h(\bar{y}))$ is calculated using:

$$w_h(\bar{y}) = 100 \cdot \left(\frac{u(\bar{y})}{\bar{y}}\right) \quad \text{(A29)}$$

Where $\bar{y}$ is the calculated mean of the automatic daily mean concentrations.

The expanded uncertainty in the mean is calculated using a coverage factor of $k = 2$ reflecting a 95% confidence interval with a normal distribution associated with the large number of measurements. Therefore:

$$W(\bar{y}) = k \cdot w_h(\bar{y}) = 2w_h(\bar{y}) \quad \text{(A30)}$$

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