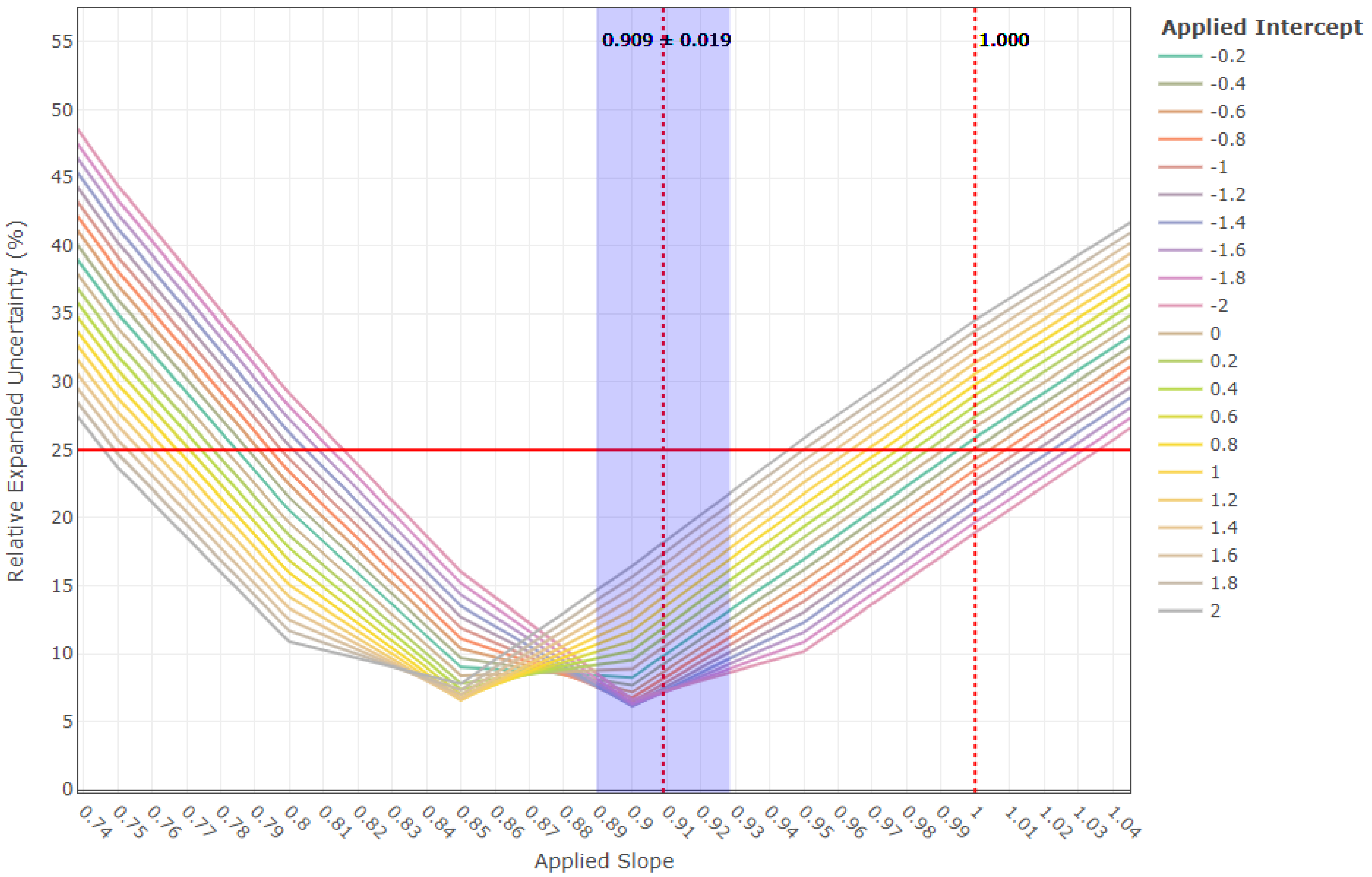

EQUIVALENCE STUDY TO INVESTIGATE PARTICULATE MATTER MONITORING IN SCOTLAND USING THE FIDAS 200

Report for: Scottish Government

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Ricardo is certified to ISO9001, ISO14001, ISO27001 and ISO45001.

Executive summary

Ricardo Energy & Environment (Ricardo) was contracted by Scottish Government to investigate the relationship between automatic particulate matter (PM_{10} and $PM_{2.5}$, collectively referred to as PM) measurement techniques used in Scotland and the EU reference method in 2020. This study is a review of PM_{10} and $PM_{2.5}$ data collected between January 2020 and June 2022 and builds on the results from the pilot project by strengthening the monitoring methodology to more closely follow BS EN 16450 – 'Automated measuring systems for the measurement of the concentration of particulate matter'. The primary change was the introduction of duplicate reference MPNS and Fidas 200 (Method 11) automatic instruments in line with BS EN 16450, providing a more robust dataset for identifying whether it would be appropriate to apply correction factors to PM_{10} and $PM_{2.5}$ Fidas data within the SAQD.

It is important to note that these results do not supersede the equivalence designation for the Fidas 200 using the Method 11 algorithm (Fidas). The results highlight that current corrections for equivalence may not be accurately representing how the Fidas responds in Scotland's pollution and meteorological environment.

Reviewing the results for the PM_{10} comparison using data from the duplicate instruments between 08/07/2021 to 29/06/2022 indicates that the Fidas does not pass the 25% criteria for measurement uncertainty (W_{CM}) when not corrected. When the data are corrected for both slope and intercept by adding 1.993 then dividing by 0.909 the Fidas meets the requirement for measurement uncertainty with a W_{CM} of 7.3% using all data. Simplifying the correction by dividing by 0.909 also meets this requirement with a W_{CM} of 11.0%. The following table summarises the PM_{10} results:

Instrument	PM_{10} Measurement Uncertainty at 50 $\mu g\ m^{-3}$		
	No correction	Correction by dividing by adding 1.993 then 0.909	Correction by dividing by 0.909
Fidas 200	26.5%	7.3%	11.0%

Reviewing the results for the $PM_{2.5}$ comparison using data from the duplicate instruments between 08/07/2021 to 29/06/2022 indicates that the Fidas passes the 25% criterion with a W_{CM} of 10.7%. This is also the case for data corrected for intercept by adding 1.162 with a W_{CM} of 12.7% using all data. Using the current correction of gravimetric equivalence, 1.06, W_{CM} is 16.5%. These results therefore indicate that no correction would be the most appropriate approach within the SAQD. The following table summarises the $PM_{2.5}$ results:

Instrument	$PM_{2.5}$ Measurement Uncertainty at 30 $\mu g\ m^{-3}$		
	No correction	Correction by adding 1.162	Correction by dividing by 1.06
Fidas 200	10.7%	12.7%	16.5%

Considering the results of this study, the following recommendations are made to the Scottish Government:

- As this study does not supersede the formal UK equivalence results, the corrections for gravimetric equivalence currently applied on the Scottish Air Quality Database and website[1] will remain unchanged.
- Fidas PM_{10} data collected within the SAQD should be corrected by dividing by 0.909, which should be applied to 2022 data onwards. A correction for slope only is recommended for simplicity and authorities will need to apply this correction to data downloaded from the Air Quality in Scotland website.
- Fidas $PM_{2.5}$ data collected within the SAQD should not be corrected. This will mean that authorities will need to apply a correction to data downloaded from the Air Quality in Scotland website by multiplying 1.06 applied to 2022 data onwards.
- For completeness, it is recommended that authorities report both the corrected, by applying the corrections defined above, and uncorrected results, as reported on the Air Quality in Scotland website within their reports.

[1] https://www.scottishairquality.scot/

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1. INTRODUCTION

Ricardo Energy & Environment (Ricardo) was contracted by Scottish Government to investigate the relationship between automatic particulate matter (PM_{10} and $PM_{2.5}$, collectively referred to as PM) measurement techniques used in Scotland and the EU reference method in 2020. 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. The pilot study was completed in January 2021 and this report provides the results from the project extension carried out between January 2021 and June 2022, which compares the Fidas 200 (Method 11) automatic technique, referred to as Fidas in this report, with the reference gravimetric method.

1.1 BACKGROUND

Trend analysis identified a significant drop in PM_{10} concentrations across the Scottish Air Quality Database (SAQD) network, which coincided with a change in measurement technique within the SAQD network to the Fidas; as an alternative to beta attenuation monitors (BAM) and filter dynamics measurement systems (FDMS). Similar step changes were 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 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.

Previous work 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 representative locations found that:

- The relationship between SEQ[2] (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 µg 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 µg m^{-3} difference seen with Fidas measurements.
- Research suggests that this apparent Fidas under-read is strongly correlated to black carbon concentrations and appears to be worst when the sampling inlet is less than 0.5 m from the kerb of a heavily trafficked road.

[2] 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 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 two different locations (160 days minimum), with strict requirements for the range of concentration measurements.

Ongoing equivalence is also assessed at a UK level, however, data currently available 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 southeast 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	3	2
TEOM	0	0
FDMS	0	0
Fidas	80	

Table 3 WHO air quality guidelines, 2021

Pollutant	Concentration	Measured as
$PM_{2.5}$	15 µg m^{-3} 5 µg m^{-3}	24-hour mean Annual mean
PM_{10}	45 µg m^{-3} 15 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 an ongoing equivalence research study between January 2020 and July 2022, which compares 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 STANDARD BS EN 16450

The Standard BS 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. BS 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 the Fidas instrument used within the SAQD network when measuring the lower PM concentrations typically experienced in Scotland.

1.3 SUMMARY FROM PILOT STUDY[3]

Table 4 summarises the corrections for slope for gravimetric equivalence that were identified using the data from the initial 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 Fidas analyser under-read PM_{10} concentrations by 22% and under-read $PM_{2.5}$ concentrations by 23%, compared to the reference method.

Table 4 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}	$PM_{2.5}$
Fidas	Divide by 0.817	Divide by 0.808

The calculated measurement uncertainties for PM_{10} at the limit value of 50 μg m^{-3} for the Fidas before and after correction for slope are provided in Table 5. The data indicated that the Fidas did not meet the ±25% threshold for equivalence without correction but met this requirement once corrected.

Table 5 PM_{10} measurement uncertainty of daily average concentrations

Instrument	PM_{10} Measurement Uncertainty at 50 μg m^{-3}	
	Before Correction for Slope	After Correction for Slope
Fidas	±43.2%	±8.5%

Details of the calculated measurement uncertainties for $PM_{2.5}$ at a limit value of 30 μg m^{-3} for the Fidas before and after correction for slope are shown in Table 6. The data indicated that the Fidas did not meet the ±25% threshold for equivalence without correction for slope but did meet this requirement once corrected.

Table 6 $PM_{2.5}$ measurement uncertainty of daily average concentrations

Instrument	$PM_{2.5}$ Measurement Uncertainty at 30 μg m^{-3}	
	Before Correction for Slope	After Correction for Slope
Fidas	±41.9%	±17.9%

[3] Pilot Research Study to Investigate Particulate Matter Monitoring Techniques in Scotland, 19/08/2021: https://www.scottishairquality.scot/sites/default/files/publications/2021-09/Pilot_Research_Study_to_Investigate_PM_Monitorng_Techniques_in_Scotland_issue_1.pdf

The results therefore indicate that current corrections for equivalence may not be accurately representing how the Fidas responds at the lower concentration levels and meteorological conditions such as those observed in Scotland.

Taking into consideration the results of this study, Scottish Government decided to extend the study for a further 12 months. This extension focusses on the Fidas analyser and is more closely aligned with EN 16450, using duplicate Fidas and reference MPNS instruments at the Glasgow Hope St monitoring site. This approach provides 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 for Fidas data were 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.

2. METHODOLOGY

2.1 MEASUREMENT TECHNIQUE INVESTIGATED

As part of this study, the following measurement technique was assessed against an EU reference method that meets the requirements of EN 12341[4] (MicroPNS (MPNS) Type HVS16 gravimetric sampler[5]):

- Fidas[6] 200 with the current approved algorithm for converting particle numbers to mass concentrations (Method 11).

The Fidas has 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 7 details the corrections for slope required to meet the equivalence criteria for PM_{10} and $PM_{2.5}$.

Table 7 Corrections required for equivalence for PM_{10} and $PM_{2.5}$, Fidas 200 (Method 11)

Analyser Type	Correction for Gravimetric Equivalence	
	PM_{10}	$PM_{2.5}$
Fidas (Method 11)	No correction	Divide by 1.060

2.2 SAMPLING REGIME

Monitoring of both PM_{10} and $PM_{2.5}$ is being carried out at Glasgow Kerbside (Hope St, Glasgow - https://www.scottishairquality.scot/latest/site-info/GLA4) between January 2020 and July 2022. 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. 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, the MPNS PM_{10} sample heads were swapped with a $PM_{2.5}$ heads to measure the two size fractions on a 4-weekly rotation.

[4] 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

[5] https://www.mcz.de/umwelttechnik_23_Low-Volume-Sampler-LVS16_en.php

[6] https://www.palas.de/en/product/fidas200s

Figure 1 shows the site configuration with the location of the Fidas automatic analysers and MPNS reference samplers within the monitoring site and relative to Hope Street, Glasgow. For context, a 3-dimensional representation of the site location is also provided in Figure 2. Fidas-2 was installed on 20/05/2021 and MPNS-2 on 08/07/2021.

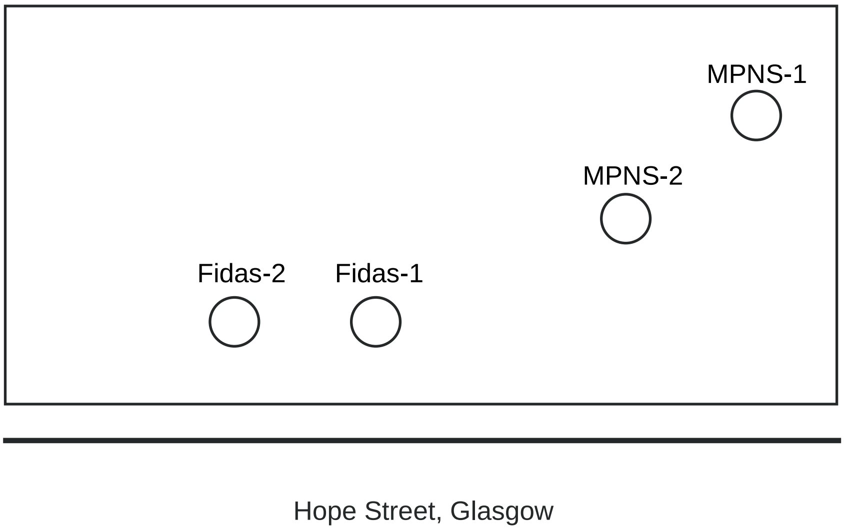


Figure 1 Glasgow Kerbside AURN monitoring site configuration (not to scale)

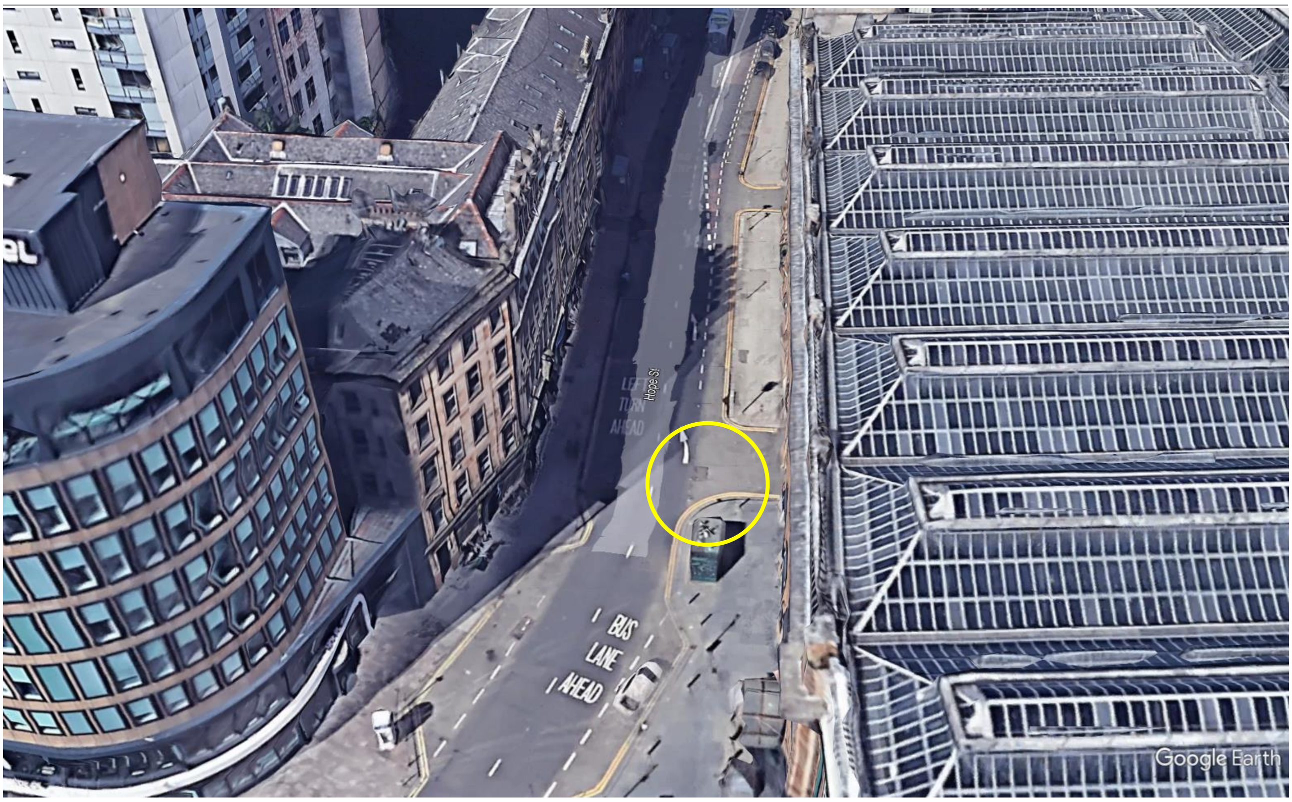


Figure 2 Location of Glasgow Kerbside (coordinates: 55.859170, -4.258889), Hope Street, Glasgow

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

To assess the performance of the MPNS sampler and Fidas instruments, 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. 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.

- 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.

2.3.2 MPNS filter handling and weighing

The MPNS sampler samples ambient air through a filter daily and 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

The following are closely examined during the data ratification process:

- Issues that have been flagged up automatically by the software or during the daily checks
- Comparison with other monitoring sites
- General review of the result to make sure that there are no other anomalies.

Once the data are 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.

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[7]. Identified outliers were then removed from the dataset and the regression analysis was carried out again.

3. RESULTS

3.1 MONITORING RESULTS

The Scottish Government implemented lockdown measures in March 2020 because of the COVID-19 pandemic. As a consequence, PM_{10} and $PM_{2.5}$ concentrations declined[8] compared to what would normally be expected. Although the improvement in air quality was a welcome consequence of the lockdown measures, the decrease in PM concentrations compounded the uncertainty in the pilot study results (16/01/2021 – 13/01/2021). The main reason for this is due to the increased noise in the reference sampler datasets as PM concentrations approach the limit of detection (LoD) of the technique.

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 (as measured by the reference method). This range in concentrations is very hard to achieve so it is common practice to perform analysis once 10 measurements above the threshold are obtained. During the pilot 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. In comparison, nine and three days greater than 28 and 17 µg m^{-3} were measured during the extended study, respectively (08/07/2021 – 29/06/2022).

Average PM concentrations measured during the pilot and extended studies are detailed in Table 8 below – note that the average concentrations were calculated using daily averages when both samplers/analysers were operating. As might be expected, average concentrations increased during 08/07/2021 to 29/06/2022 when compared to the pilot study with the relaxation of lockdown measures. However, the increase as measured by the reference MPNS sampler and Fidas differed in magnitude with the MPNS measuring an increase of 3.2% and 4.8% in PM_{10} and $PM_{2.5}$ concentrations, respectively, compared to 15.7% and 15.1% measured by the Fidas.

Figure 3 and Figure 4 show the range of PM concentrations measured each year from January 2020 to June 2022. Again, these plots illustrate how PM concentrations increased as the lockdown measures were relaxed with a noticeable increase in 2022 at the time when measures were fully removed. This increase can be seen in both the range of concentrations measured by both methods and by the increase in maximum PM concentrations. Specifically, for $PM_{2.5}$ during 2022 the maximum daily mean concentrations measured by the Fidas instruments were significantly greater than that seen by the MPNS. This is reflected in the number of days greater than 17 µg m^{-3} with 14 days measured by the Fidas compared to three by the MPNS.

In addition to the increase in concentrations from year to year, average PM_{10} and $PM_{2.5}$ concentrations measured by the MPNS sampler were up to 38% and 21% greater than that measure by the Fidas analyser. This difference together with the difference in increase in concentrations highlights and re-emphasises the

[7] Generalized ESD Test for Outliers: https://www.itl.nist.gov/div898/handbook/eda/section3/eda35h3.htm
[8] Impact of Lockdown Measures on Scottish Air Quality in 2020, 14/07/2021: https://www.scottishairquality.scot/sites/default/files/publications/2021-07/SAQD_Covid19_Technical_Report_Issue_1.pdf

fundamental differences between the reference method and Fidas measurement techniques (gravimetric compared to optical technique).

There are also differences in measurements between the duplicate MPNS and Fidas instruments. For example, average PM_{10} concentrations measured by MPNS-1 are greater than MPNS2 and Fidas-1 greater than Fidas-2; and for $PM_{2.5}$, MPNS-1 is greater than MPNS2 and Fidas-1 greater than Fidas-2. This difference is also reflected in the 25 to 75 quartile range with MPNS-1 and Fidas-1 also measuring a wider range of concentrations when compared to MPNS-2 and Fidas-2, respectively. This highlights the underlying uncertainties in the PM measurements even between samplers/ analysers of the same type.

Table 8 Average PM concentrations measured during the pilot and extended studies

Analyser	PM_{10}		$PM_{2.5}$	
	16/01/2021 - 07/07/2021	08/07/2021 – 29/06/2022	16/01/2021 - 07/07/2021	08/07/2021 – 29/06/2022
MPNS-1	15.7	16.3	8.4	9.2
MPNS-2	-	15.8	-	8.5
Fidas-1	11.4	13.3	6.6	7.8
Fidas-2	-	12.0	-	7.3

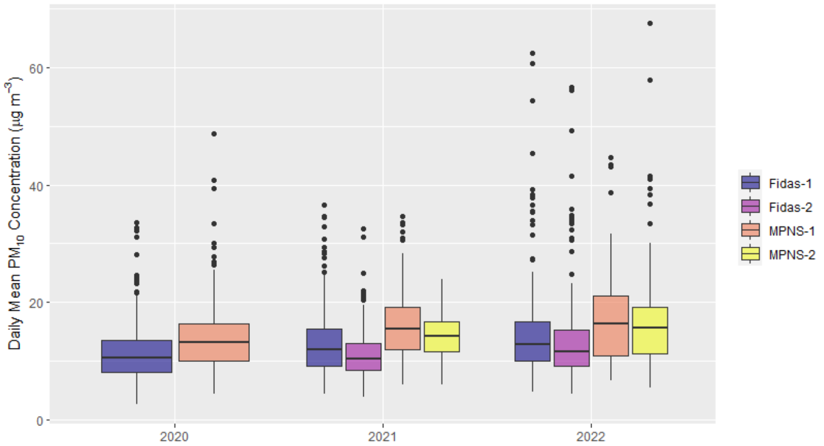


Figure 3 Box plot of annual PM_{10} concentrations, 2020 - 2022

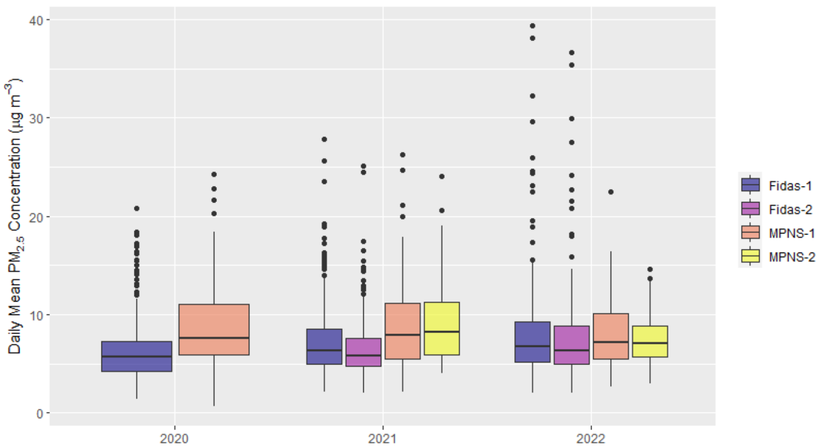


Figure 4 Box plot of annual $PM_{2.5}$ concentrations, 2020 - 2022

3.2 RESULTS FROM DUPLICATE MPNS AND FIDAS

Looking more closely at the intra-relationships between the duplicate MPNS samplers, Figure 5 and Figure 6 show the comparison of daily PM_{10} and $PM_{2.5}$ concentrations between MPNS-1 and MPN-2 samplers in the form of time series and scatter plots. The plots highlight again the differences between the two samplers with PM_{10} and $PM_{2.5}$ concentrations approximately 9% and 12% greater as measured by MPNS-1 compared to MPNS-2. The correlation in concentrations between both samplers is strong with R^2 of 0.97 and 0.93, respectively, with the weaker correlation for $PM_{2.5}$ is reflective of the lower concentrations measured and the associated increase in noise closer to the LoD.

The site location within a street canyon and inlet locations might have an impact on the measured data, as there could be a concentration gradient due to complex air flow within the canyon. A further contribution to the difference seen is the intrinsic uncertainty associated with the measurements. The key requirement, however, is that the between reference method uncertainty should be less than 2.0 µg m^{-3}, which is achieved.

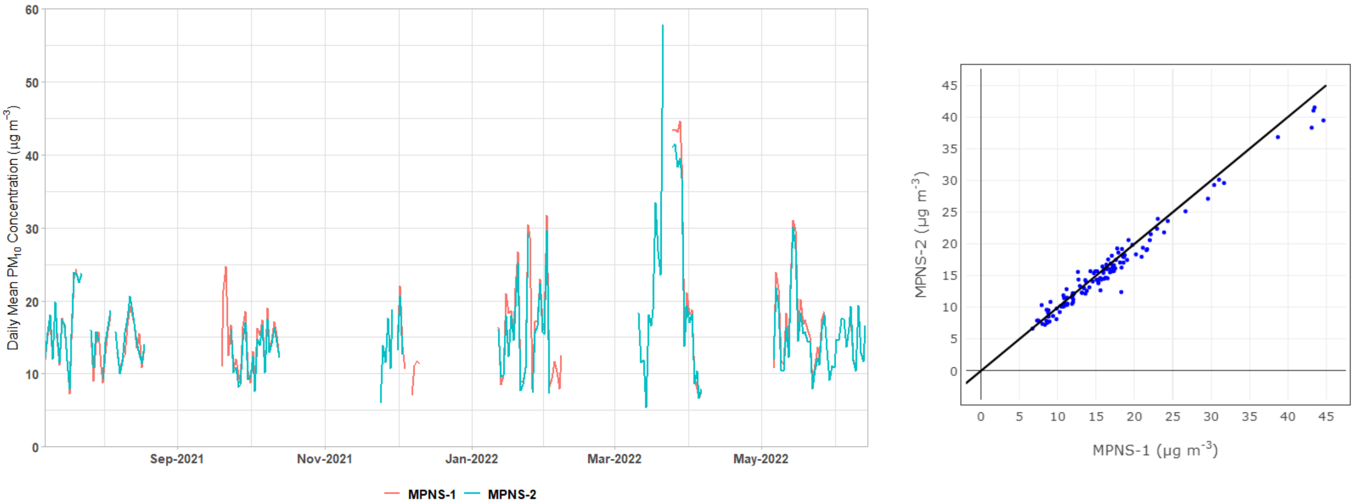


Figure 5 Comparison plots of duplicate MPNS daily mean PM_{10} concentrations

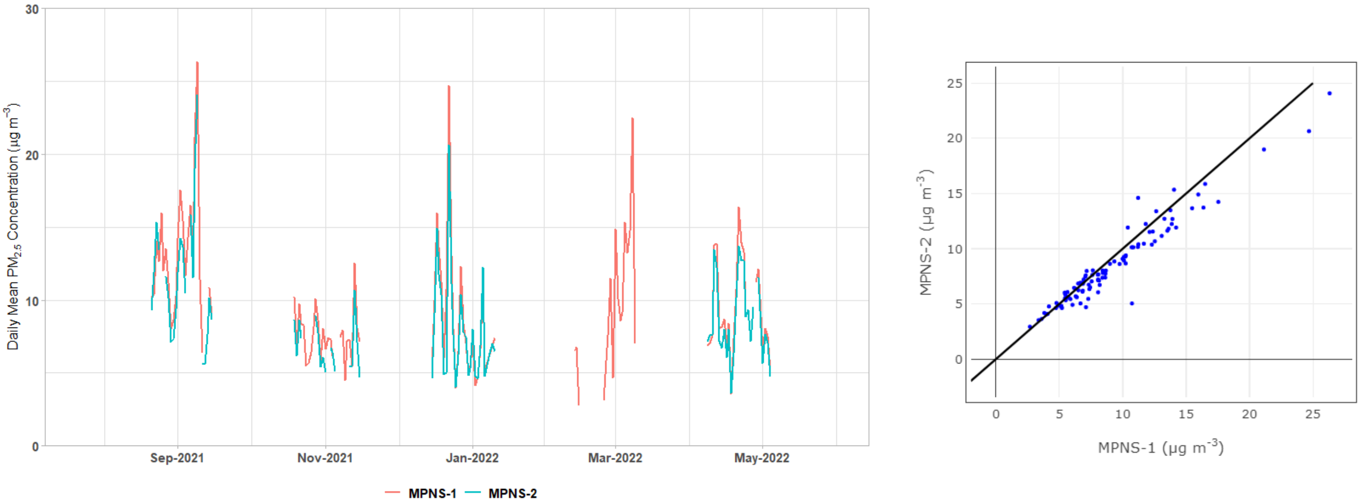
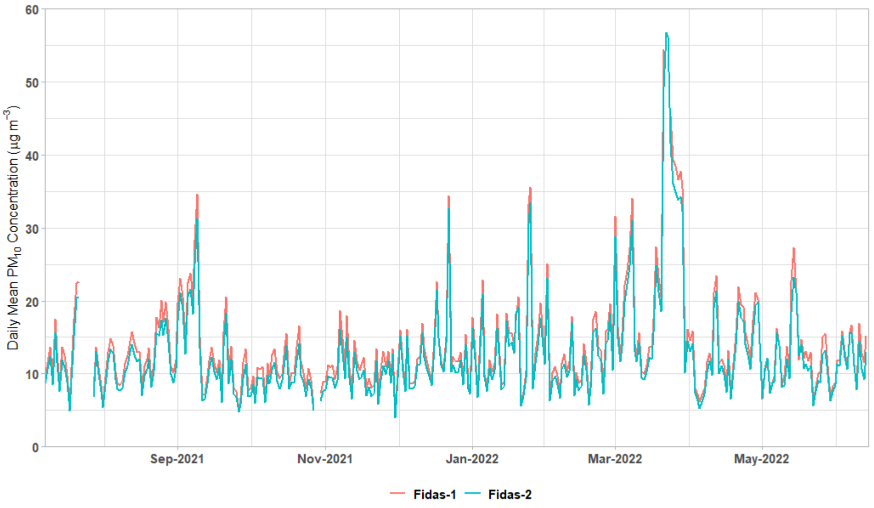


Figure 6 Comparison plots of duplicate MPNS daily mean $PM_{2.5}$ concentrations

The comparison in PM_{10} and $PM_{2.5}$ concentrations between duplicate Fidas analysers is shown in Figure 7 and Figure 8, respectively. Again, a difference can be seen in PM concentrations measured by the duplicate analysers with Fidas-1 measuring PM concentrations of approximately 9% and 7% higher than Fidas-2. The correlation between the two Fidas is stronger than that seen in the MPNS measurements with R^2 of greater than 0.99 for both PM_{10} and $PM_{2.5}$. This improved correlation is likely due in part to a lower LoD associated with the Fidas.

Again, there will be factors such as the site location within a street canyon and intrinsic measurement uncertainty that will likely be contributing factors in the difference seen. The key requirement that the between candidate method uncertainty should be less than 2.5 µg m^{-3} is achieved.



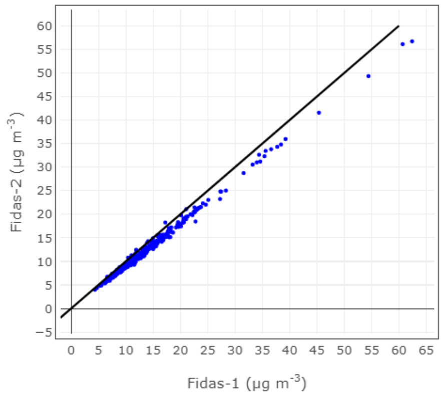
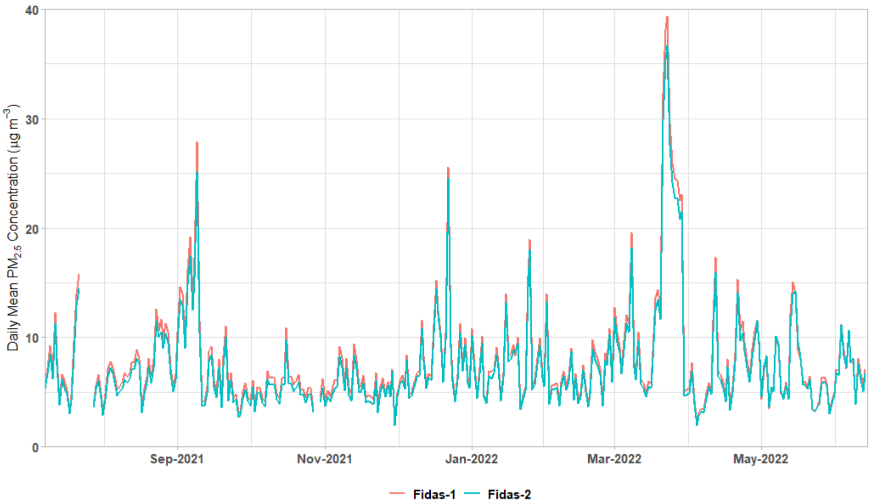


Figure 7 Comparison plots of duplicate Fidas daily mean PM_{10} concentrations



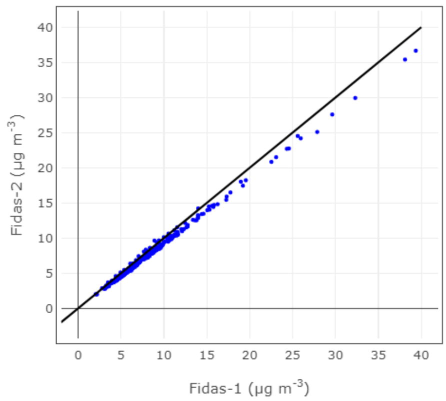


Figure 8 Comparison plots of duplicate Fidas daily mean $PM_{2.5}$ concentrations

3.3 RESULTS FROM PM_{10} COMPARISON

The following section details the results from the comparison between the MPNS reference samplers and Fidas daily PM_{10} measurements for the period 08/07/2021 to 29/06/2022. The average of the duplicate MPNS and Fidas instruments are used as the comparison datasets and are calculated only when all instruments were operating.

Table 9 details the following results of the regression analysis for PM_{10} for five datasets:

- Winter comparison using data between October and March
- Summer comparison using data between April and September
- A comparison using data when reference measurements are greater or equal to 30 µg m^{-3}
- A comparison using data when reference measurements are less or equal to 30 µg m^{-3}
- A comparison using all data

In total 117 data pairs (n) were collected between 08/07/2021 to 29/06/2022 with all datasets showing strong correlation with $R^2 > 0.8$. Only seven (7.7%) daily averages recorded by the reference sampler were greater than 28 µg m^{-3}, however, this reflects typical PM_{10} concentrations measured across Scotland. Of the five datasets, only one indicated a relative expanded measurement uncertainty (W_{CM}) of less than 25% for uncorrected Fidas data ($PM_{10} \geq 30$ µg m^{-3}).

Taking all data together, no outliers were identified using the ESD test, and the results indicate that the Fidas PM_{10} data have a measurement uncertainty of 26.5% and require a correction for both slope and intercept. The corresponding scatter plot is shown in Figure 9 - the between MPNS (u(bs,RM)) and Fidas (u(bs,CM)) uncertainty from the duplicate measurements meet the requirement of 2.0 and 2.5 µg m^{-3}, respectively.

Table 9 Regression results from comparison of daily PM_{10} concentrations (08/07/2021 to 29/06/2022)

Glasgow Hope Street	Dataset	Orthogonal Regression – PM_{10}								LV of 50 µg m^{-3}	
		n	R^2	Slope (b)	±	ub	Intercept (a)	±	ua	W_{cm}	%>28
MPNS vs Fidas	Winter	45	0.96	0.923	±	0.028	-1.964	±	0.585	24.06	15.6
	Summer	72	0.91	0.840	±	0.030	-1.142	±	0.464	36.64	2.8
	≥30	7	0.98	1.049	±	0.074	-7.286	±	2.839	19.26	100
	<30	110	0.87	0.894	±	0.032	-1.774	±	0.486	28.61	0
	All data	117	0.95	0.909	±	0.019	-1.993	±	0.342	26.48	7.7

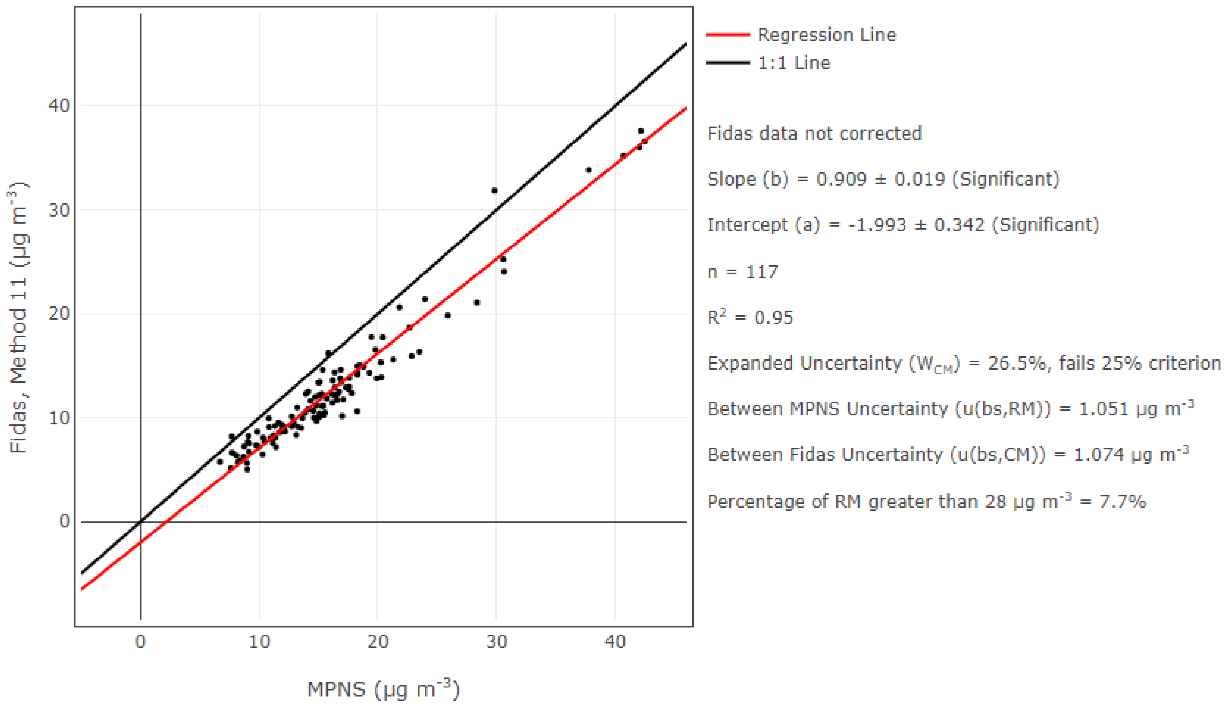


Figure 9 Scatter plot of PM_{10} concentrations – Fidas data not corrected

The results of applying both a slope and intercept correction to the Fidas PM_{10} data are provided in Table 10, which provides the following:

- A comparison using data when reference measurements are greater or equal to 30 µg m^{-3}
- A comparison using data when reference measurements are less or equal to 30 µg m^{-3}
- A comparison using all data
- The between sampler uncertainties for each dataset

The results indicate that when the data are corrected for both slope and intercept the Fidas meets the requirement for measurement uncertainty with W_{CM} well below 25% using all datasets. The between reference sampler uncertainty is greater than 2.0 µg m^{-3} and candidate method uncertainty is greater than 2.5 µg m^{-3} using data greater than 30 µg m^{-3}, however, this again is likely due to in part to the low number of days above 30 µg m^{-3} (seven). Using all data, W_{CM} drops to 7.25% when the Fidas data are corrected for slope and intercept by adding 1.993 then dividing by 0.909 (Figure 10).

Table 10 Regression results from comparison of PM_{10} Fidas concentrations corrected for slope and intercept

CM corrected by adding 1.993 then dividing by 0.909	7.7% > 28 µg m^{-3}	Orthogonal Regression – PM_{10}								Between Instrument Uncertainties (µg m^{-3})	
	W_{cm} (%)	n	R^2	Slope (b)	±	ub	Intercept (a)	±	ua	MPNS (u(bs,RM))	Fidas (u(bs,CM))
≥30	9.20	7	0.98	1.156	±	0.081	-5.876	±	3.122	2.193	2.522
<30	7.56	110	0.87	0.991	±	0.034	0.139	±	0.535	0.932	1.042
All data	7.25	117	0.95	1.003	±	0.021	-0.041	±	0.376	1.051	1.184

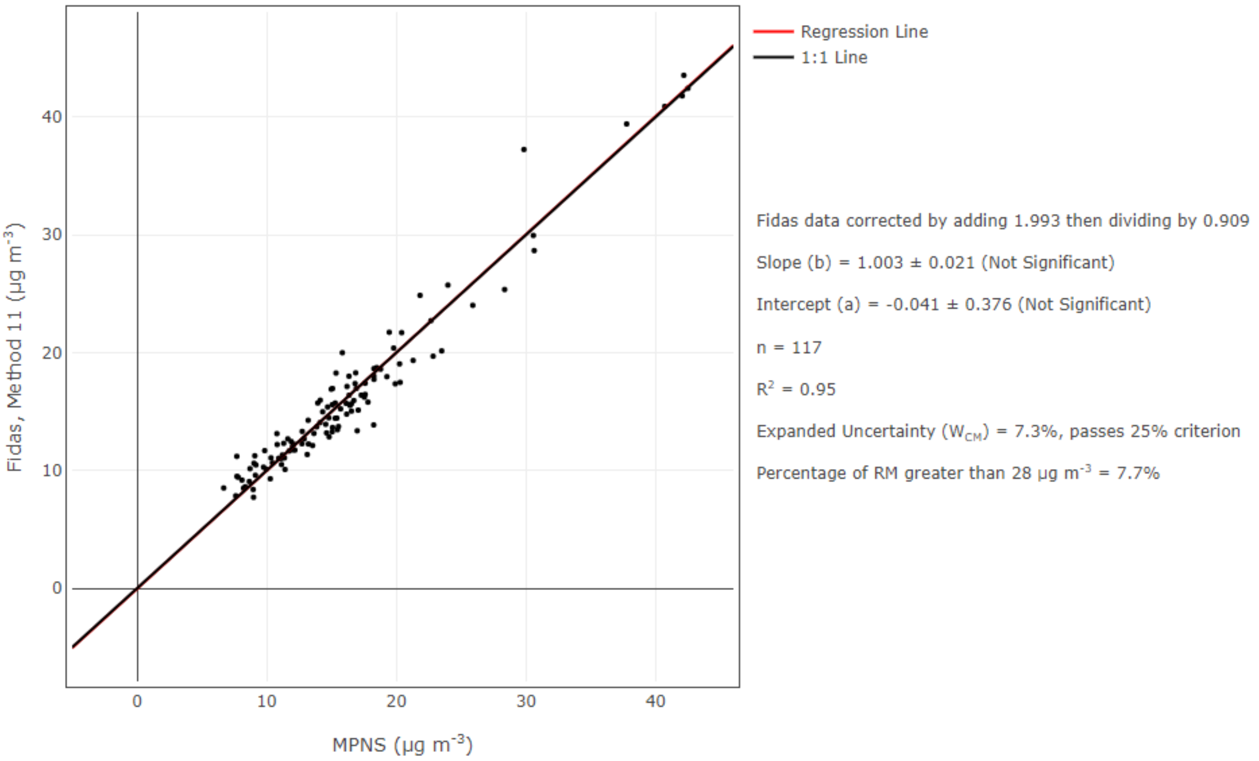


Figure 10 Scatter plot of PM_{10} concentrations – Fidas data corrected by adding 1.993 and then dividing by 0.909

It is also appropriate to investigate whether applying a slope correction only improves the measurement uncertainty of the Fidas data. Table 11 and Figure 11 detail the results from the comparison of Fidas data corrected for slope by dividing by 0.909, and the MPNS. Using all data W_{CM} is 11.0% and so the Fidas PM_{10} meets the requirement of 25% for PM_{10} when corrected for slope only.

Table 11 Regression results from comparison of PM_{10} Fidas concentrations corrected for slope only

CM corrected by dividing by 0.909	7.7% > 28 µg m^{-3}	Orthogonal Regression – PM_{10}								Between Instrument Uncertainties (µg m^{-3})	
	W_{cm} (%)	n	R^2	Slope (b)	±	ub	Intercept (a)	±	ua	MPNS (u(bs,RM))	Fidas (u(bs,CM))
≥30	5.01	7	0.98	1.156	±	0.081	-8.068	±	3.123	2.193	2.522
<30	12.37	110	0.87	0.991	±	0.035	-2.053	±	0.535	0.932	1.042
All data	11.0	117	0.95	1.003	±	0.021	-2.234	±	0.376	1.051	1.184

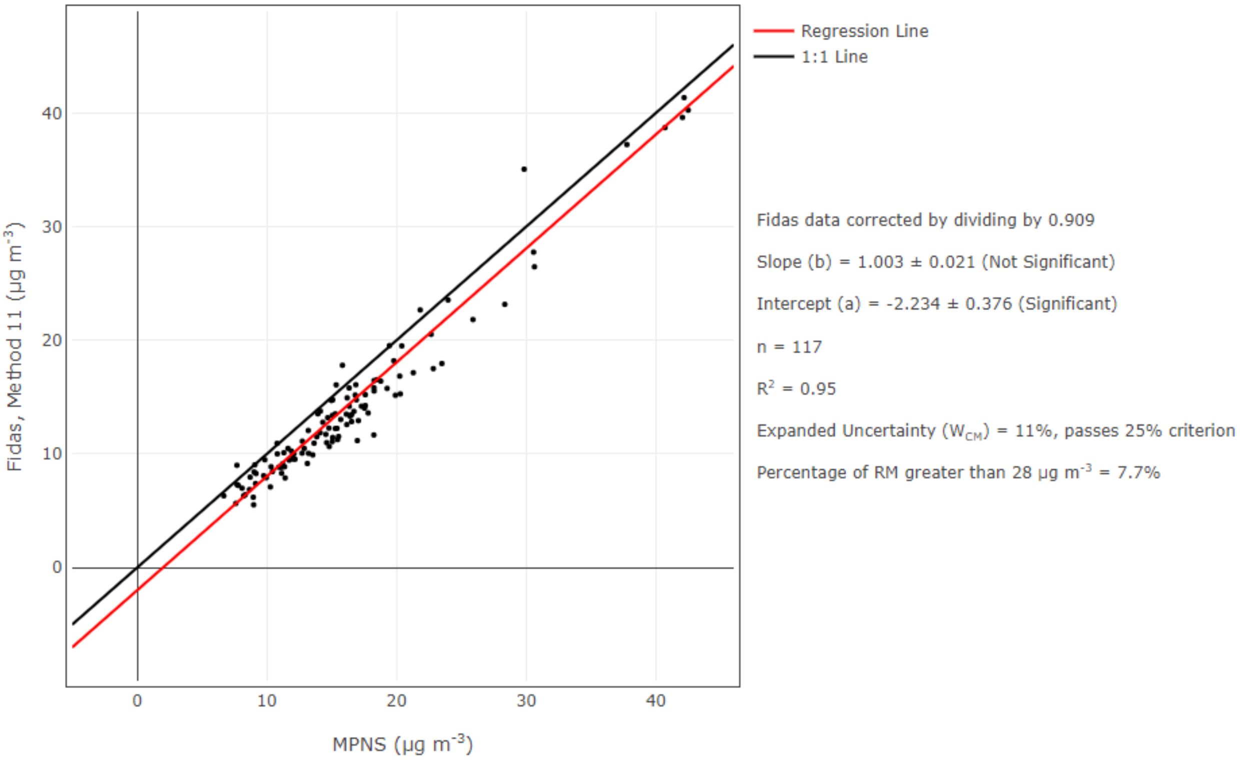


Figure 11 Scatter plot of PM_{10} concentrations – Fidas data corrected by dividing by 0.909

3.4 RESULTS FROM $PM_{2.5}$ COMPARISON

The following section details the results from the comparison between the MPNS reference samplers and Fidas daily $PM_{2.5}$ measurements for the period 08/07/2021 to 29/06/2022 and without using the current correction for gravimetric equivalence by dividing by 1.06 (Table 12). The average of the duplicate MPNS and Fidas instruments are used as the comparison datasets and are calculated only when all instruments were operating. Table 12 provides the following:

- Winter comparison using data between October and March
- Summer comparison using data between April and September
- A comparison using data when reference measurements are greater or equal to 18 µg m^{-3}
- A comparison using data when reference measurements are less or equal to 18 µg m^{-3}
- A comparison using all data

In total 97 data pairs were collected between 08/07/2021 to 29/06/2022 with all datasets showing moderately strong to strong correlation with $R^2 > 0.75$. Only three daily averages (3.1%) recorded by the reference sampler were greater than 17 µg m^{-3}, however, this again reflects typical $PM_{2.5}$ concentrations measured across Scotland. Of the five datasets, only one indicated a relative expanded measurement uncertainty (W_{CM}) of greater than 25% for uncorrected Fidas data ($PM_{2.5} \geq 18$ µg m^{-3}).

Taking all data together, no outliers were identified using the ESD test, and the results indicate that the Fidas $PM_{2.5}$ data have a measurement uncertainty of 10.66% and so results indicate that no correction for slope or intercept is required, although, the intercept is identified as being significant. The corresponding scatter plot is shown in Figure 12 - the between MPNS (u(bs,RM)) and Fidas (u(bs,CM)) uncertainty from the duplicate measurements meet the requirement of 2.0 and 2.5 µg m^{-3}, respectively.

Table 12 Regression results from comparison of daily $PM_{2.5}$ concentrations (08/07/2021 to 29/06/2022)

Glasgow Hope Street	Dataset	Orthogonal Regression – $PM_{2.5}$								LV of 30 µg m^{-3}	
		n	R^2	Slope (b)	±	ub	Intercept (a)	±	ua	W_{cm}	%>17
MPNS vs Fidas	Winter	41	0.81	1.093	±	0.075	-1.187	±	0.650	15.02	2.4
	Summer	56	0.86	1.027	±	0.052	-1.443	±	0.544	11.26	3.6
	≥18	3	0.86	2.244	±	0.801	-28.181	±	18.220	64.13	100
	<18	94	0.76	0.984	±	0.051	-0.763	±	0.457	13.03	0
	All data	97	0.84	1.033	±	0.042	-1.162	±	0.412	10.66	3.1

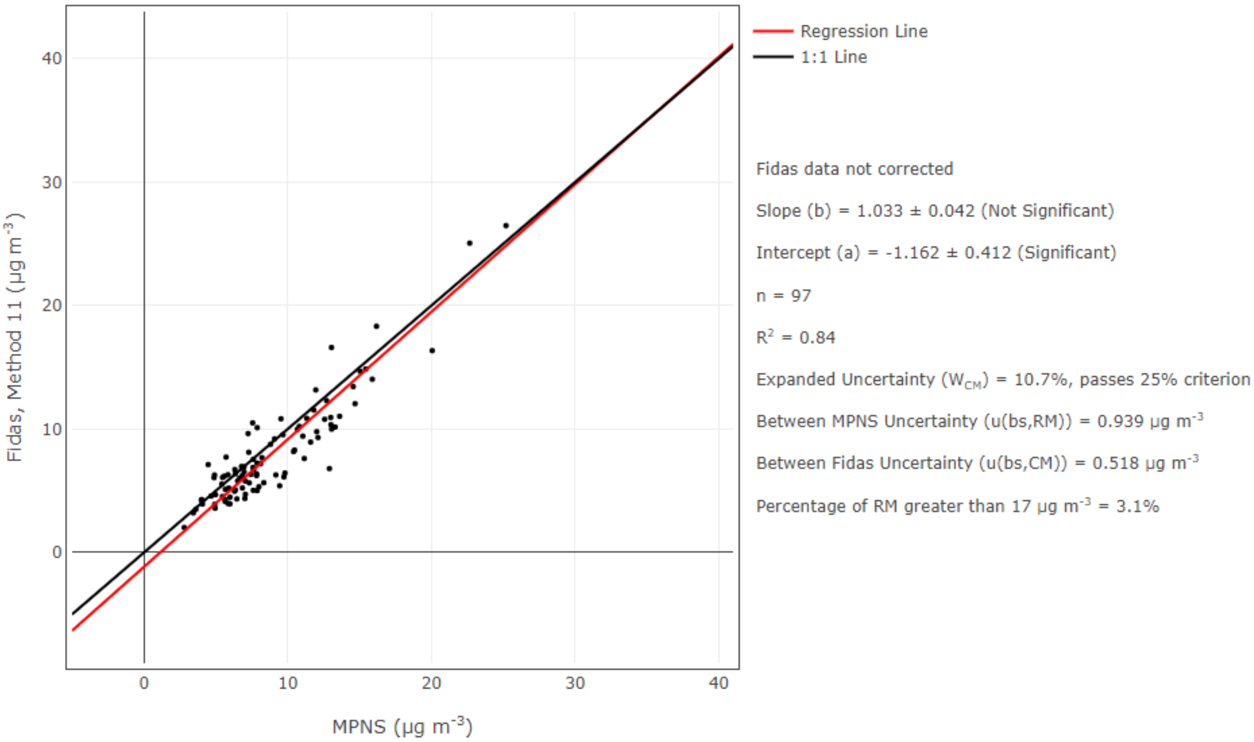


Figure 12 Scatter plot of $PM_{2.5}$ concentrations – Fidas data not corrected

The between reference sampler uncertainty was greater than 2.0 µg m^{-3} using data greater than 18 µg m^{-3}, however, this is likely due to in part to the low number of days above 18 µg m^{-3}. The u(bs,CM) for daily Fidas measurements was less than 2.5 µg m^{-3} using all datasets; and u(bs,RM) for MPNS measurements meets the requirement of less than 2.0 µg m^{-3} using data less than 18 µg m^{-3} and the whole dataset.

The results in Table 13 indicate that when the $PM_{2.5}$ data are corrected by adding 1.162 the Fidas meets the requirement for measurement uncertainty using all data and data less than 18 µg m^{-3} with W_{CM} of 12.7% and 10.4%, respectively. The between reference sampler uncertainty is greater than 2.0 µg m^{-3} using data greater than 18 µg m^{-3}, however, this again is likely due to in part to the low number of days above 18 µg m^{-3} (three).

Table 13 Regression results from comparison of daily $PM_{2.5}$ concentrations corrected for intercept only

CM corrected by adding 1.162	3.1% > 17 µg m^{-3}	Orthogonal Regression – $PM_{2.5}$								Between Instrument Uncertainties (µg m^{-3})	
	W_{cm} (%)	n	R^2	Slope (b)	±	ub	Intercept (a)	±	ua	MPNS (u(bs,RM))	Fidas (u(bs,CM))
≥18	71.59	3	0.86	2.244	±	0.801	-27.019	±	18.220	2.081	1.424
<18	10.37	94	0.76	0.984	±	0.051	0.400	±	0.457	0.878	0.460
All data	12.74	97	0.84	1.033	±	0.042	0.000	±	0.412	0.939	0.518

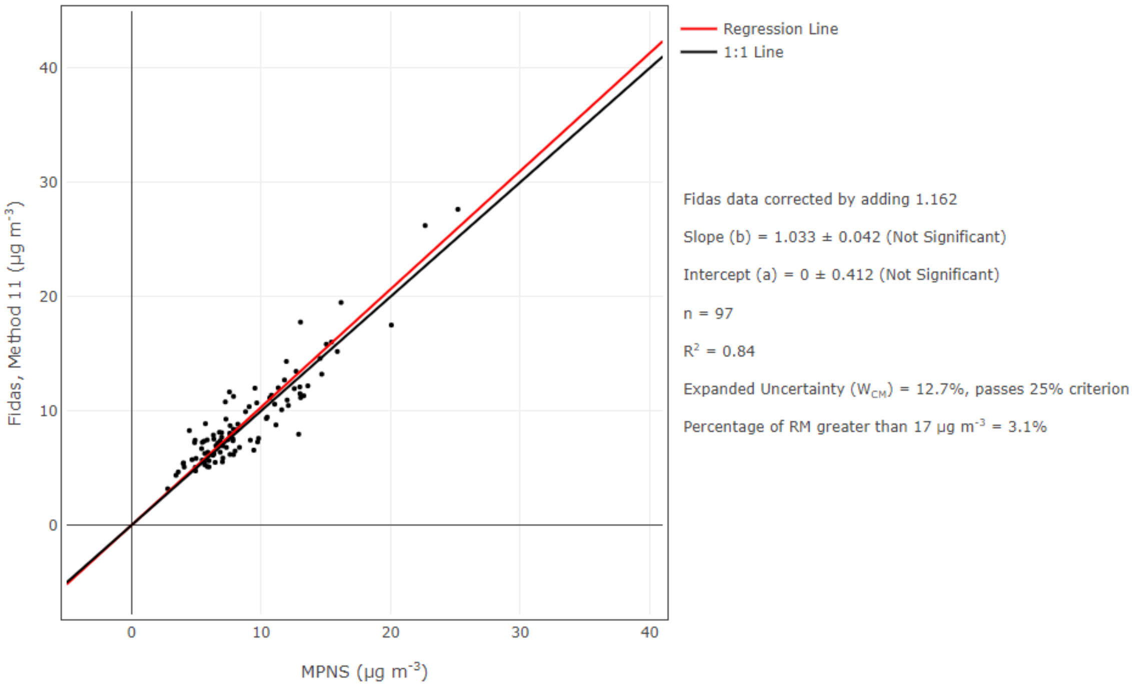


Figure 13 Scatter plot of $PM_{2.5}$ concentrations – Fidas data corrected by adding 1.162

Further analysis using $PM_{2.5}$ data corrected for gravimetric equivalence using the current correction factor of 1.06 is shown in Table 14 and Figure 14. These results are consistent with the results in Table 12 using uncorrected $PM_{2.5}$ data with the Fidas meeting the 25% requirement using all datasets except for concentrations greater than or equal to 18 µg m^{-3}. However, W_{CM} for concentrations less than 18 µg m^{-3} and using all data are greater than that for uncorrected - 13.0% and 10.7% using uncorrected data compare to 22.4% and 16.5% using concentrations divided by 1.06, respectively.

Table 14 Regression results from comparison of daily $PM_{2.5}$ concentrations corrected by dividing by 1.06

Glasgow Hope Street	Dataset	Orthogonal Regression – $PM_{2.5}$								LV of 30 µg m^{-3}	
		n	R^2	Slope (b)	±	ub	Intercept (a)	±	ua	W_{cm}	%>17
MPNS vs Fidas	Winter	41	0.81	1.025	±	0.071	-1.068	±	0.613	10.04	2.4
	Summer	56	0.86	0.965	±	0.049	-1.320	±	0.513	18.61	3.6
	≥18	3	0.86	2.111	±	0.759	-26.457	±	17.189	49.57	100
	<18	94	0.76	0.920	±	0.048	-0.652	±	0.432	22.4	0
	All data	97	0.84	0.969	±	0.040	-1.051	±	0.389	16.48	3.1

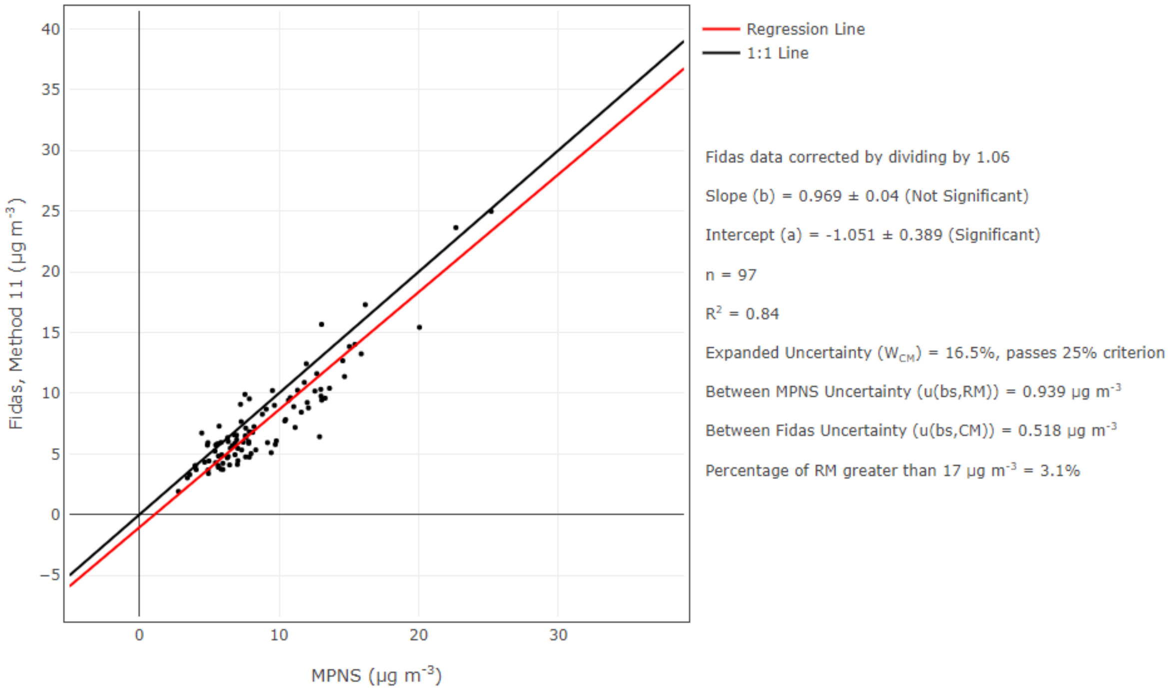


Figure 14 Scatter plot of $PM_{2.5}$ concentrations – Fidas data corrected by dividing by 1.06

4. DISCUSSION

One of the key outputs from this study is to determine whether it is appropriate to apply a correction factor to Fidas PM data within the SAQD network. The following sections, therefore, look at how the relationships between the reference MPNS samplers and Fidas analysers have varied between 2020, when the pilot study was commissioned, to 2022, which will inform whether PM_{10} and $PM_{2.5}$ Fidas data require a correction.

4.1 VARIATION IN RELATIONSHIPS – PM_{10}

Figure 15 to Figure 17, respectively, detail how the calculated intercept, slope and measurement uncertainty varied between duplicate instruments as follows: MPNS-1 compared to Fidas-1; MPNS-1 compared to Fidas-2; MPNS-2 compared to Fidas-1; and MPNS-2 compared to Fidas-2.

Also using the following datasets: Daily concentrations less than 30 µg m^{-3}; Daily concentrations greater than or equal to 30 µg m^{-3}; All data; Summer (April to September); Winter (October to March); 2020 dataset; 2021 dataset; and 2022 dataset.

Tables detailing these results are provided in Appendix 2 (Table 17 and Table 18)Figure 15 and Figure 16 demonstrate the variation in the calculated intercepts and slopes, which varied with the individual instruments that were compared, the datasets used, and with the year. There are two key observations: firstly, the calculated intercept varies greatly using daily averages of greater than or equal to 30 µg m^{-3} and when compared to the other datasets (1st quartile to 3rd quartile range: -1.4 to 5.6 µg m^{-3} compared to -1.3 to -0.5 µg m^{-3}). This increase in variation for data ≥ 30 µg m^{-3} is also seen in the calculated slope but to a lesser extent and is due to the small number of daily averages in this concentration range measured between 2020 and 2022 (19 days in total or 5.1%).

Secondly, the variation in calculated intercepts and slopes is reflected in the variation in calculated measurement uncertainties (Figure 17). The measurement uncertainty was consistently above 25% independent of dataset used and indicating that for the Scotland-specific pollution climate a correction for slope and/ or intercept is required.

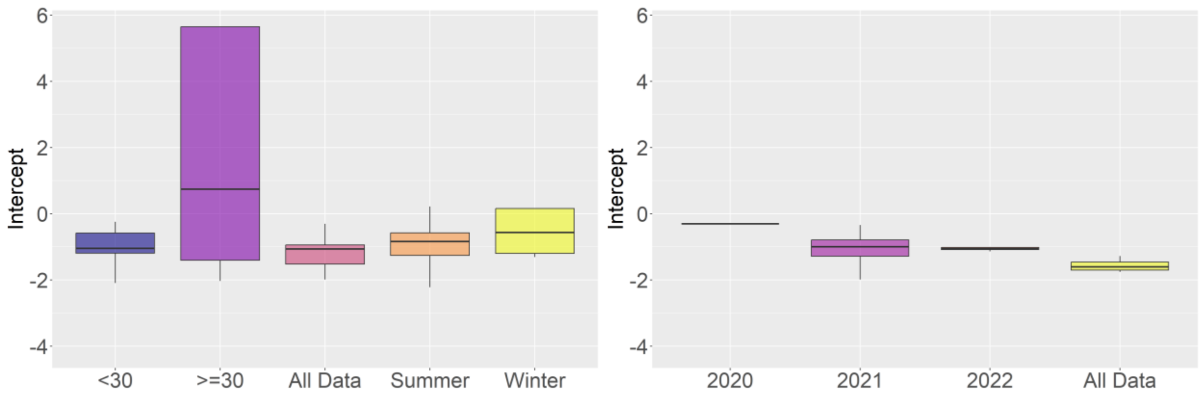


Figure 15 Variation in PM_{10} comparison intercept by data split and year

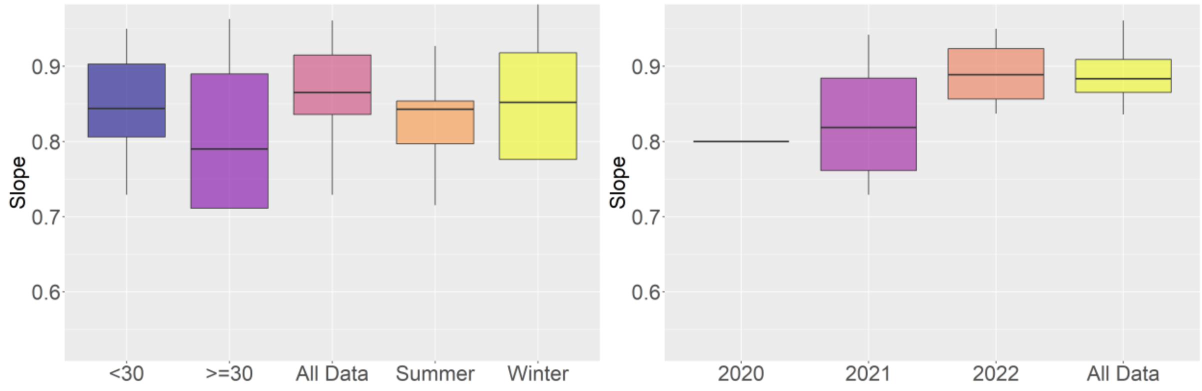


Figure 16 Variation in PM_{10} comparison uncertainty by data split and year

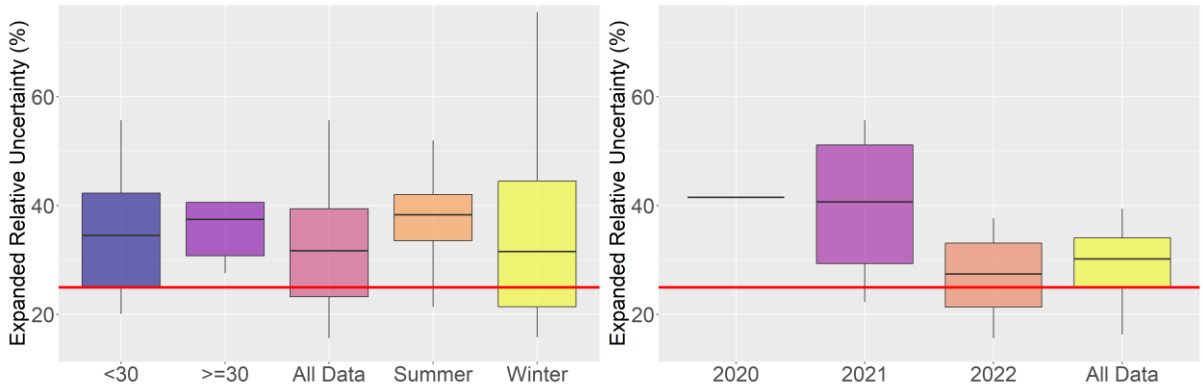


Figure 17 Variation in PM_{10} comparison uncertainty by data split and year

4.2 VARIATION IN RELATIONSHIPS – $PM_{2.5}$

A similar pattern in the variation of calculated intercept and slope is seen the $PM_{2.5}$ comparisons (Figure 18 and Figure 19). Again, the intercepts and slopes calculated using daily averages > 18 µg m^{-3} are significantly different from all other datasets, which is reflection of the low number of high data days greater than 17 µg m^{-3} measured between 2020 and 2022 (10 days or 3.2%). This large variation in intercept and slope for that dataset also feeds into the measurement uncertainty calculation and so in general measurement uncertainty is much greater than 25%. However, for all other datasets the intercepts, slopes and measurement uncertainties are much more consistent – in general, Fidas $PM_{2.5}$ measurements meet the 25% requirement without any correction to the data. This confirms that for the Scotland pollution climate, it may be appropriate to not correct Fidas $PM_{2.5}$ for slope or intercept. The higher uncertainty seen in 2020 is likely a consequence of the lower concentrations and higher noise measured.

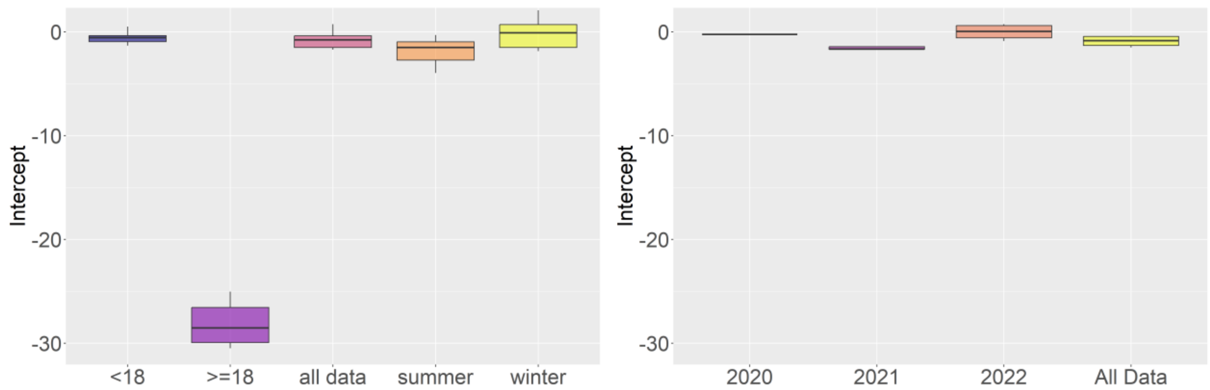


Figure 18 Variation in $PM_{2.5}$ comparison intercept by data split and year

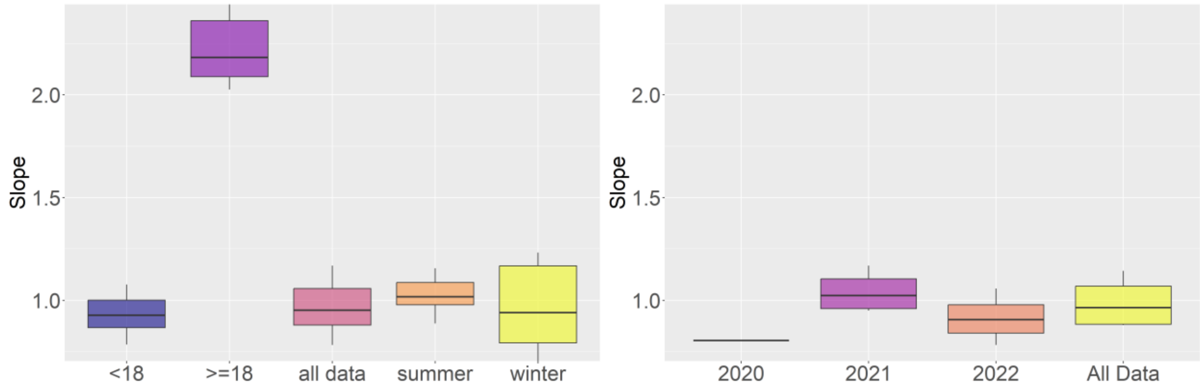


Figure 19 Variation in $PM_{2.5}$ comparison slope by data split and year

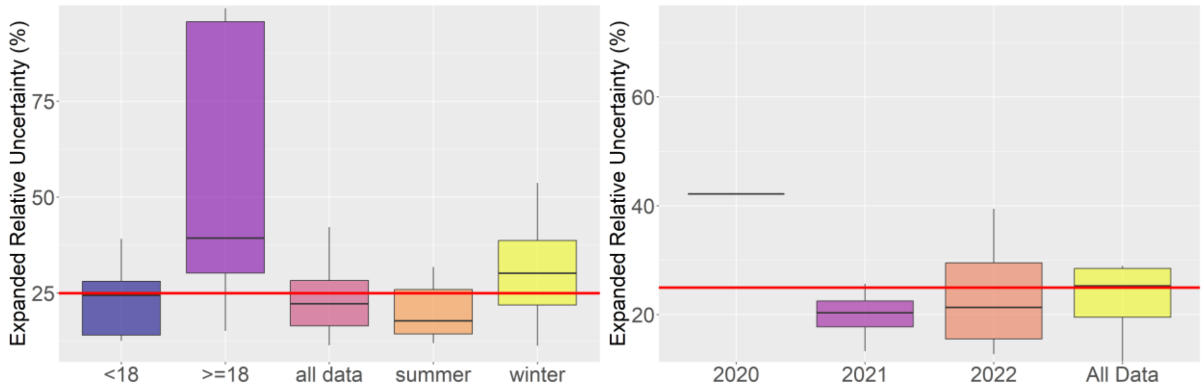


Figure 20 Variation in $PM_{2.5}$ comparison uncertainty by data split and year

4.3 VARIATION IN MEASUREMENT UNCERTAINTY

In identifying the possibility that PM_{10} and/ or $PM_{2.5}$ Fidas data require correction, Figure 21 and Figure 22 show how the relative expanded uncertainty varies with a range of intercept and slope corrections, respectively. Note that for this analysis, the uncertainty in calculated intercept (u_a) and slope (u_b) are taken from the results in Figure 9 and Figure 12 and remain constant with the variation in applied intercept and slope as follows:

- PM_{10}: u_a = 0.432, u_b = 0.019
- $PM_{2.5}$: u_a = 0.412, u_b = 0.042

Both figures show that there are a range of intercept and slope corrections that result in PM_{10} and $PM_{2.5}$ Fidas measurements meeting the 25% uncertainty requirement, but that measurement uncertainty can be optimised by selecting the most appropriate corrections. Specifically, for PM_{10} Fidas measurements, the results in Section 3.3 indicate that a correction for intercept and slope by adding 1.993 then dividing by 0.909 would be optimal and Figure 21 confirms that this correction does, as expected, minimise the measurement uncertainty. The results also indicate a second option, however, by dividing by 0.909 only, which would simplify the correction of Fidas PM_{10} data and would keep the uncertainty well below the required 25% at 11.0%. It is clear again that using the available data collected for this study that not applying a correction factor to Fidas PM_{10} data results in a failure of the 25% criterion with the 1.000 falling well outside of the uncertainty band for the calculated slope (u_b = 0.019).

Fidas $PM_{2.5}$ data show a similar pattern in Figure 22 indicating that not correcting $PM_{2.5}$ data would optimise measurement uncertainty and that using the current correction by dividing by 1.06 still meets the 25% criterion but increases the measurement uncertainty from 10.7% to 16.5%. Using either 1.00 or 1.06 falls within the uncertainty band of the calculated slope of 1.033 ± 0.042 but the improvement in measurement uncertainty at 1.000 indicates that applying no correction would be more appropriate.

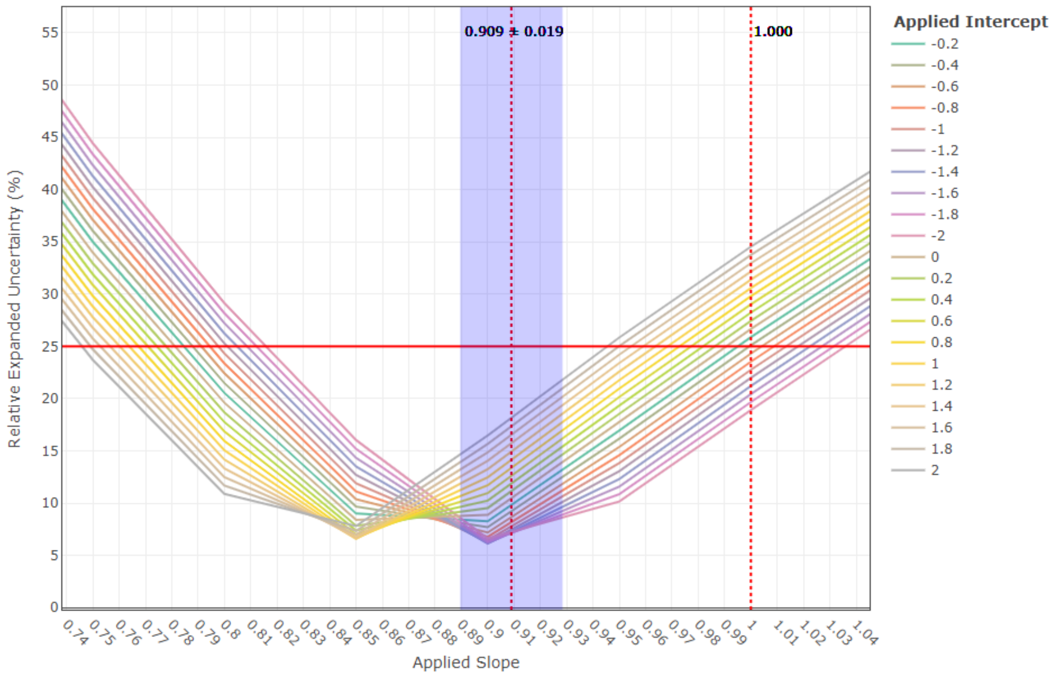


Figure 21 Relative expanded uncertainty in Fidas PM_{10} measurements with varying intercept and slope

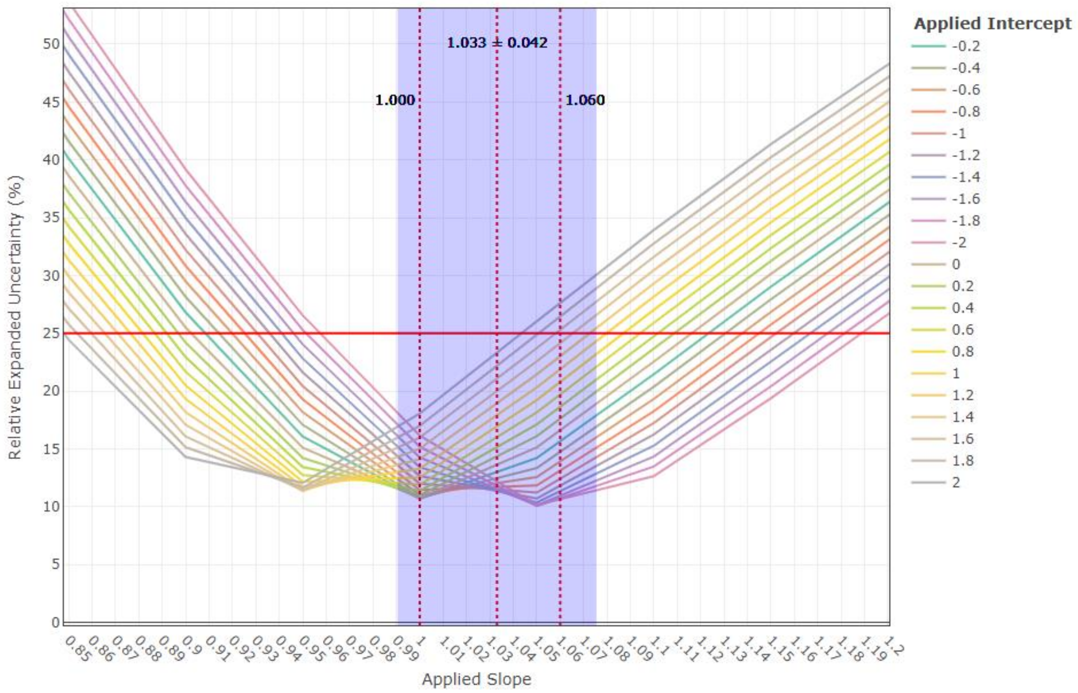


Figure 22 Relative expanded uncertainty in Fidas $PM_{2.5}$ measurements with varying intercept and slope

5. CONCLUSIONS

It is important to note that the results in this report do not supersede the equivalence designation for the Fidas 200 using the Method 11 algorithm (Fidas). The results highlight that current corrections for equivalence may not be accurately representing how the Fidas responds in Scotland's pollution and meteorological environment.

A pilot study was initially commissioned by the Scottish Government due to an identified step change decrease in particulate matter (PM_{10}) concentrations across Scotland which coincided with the introduction of Fidas within the Scottish Air Quality Database (SAQD) monitoring network. The Fidas replaced tapered element oscillating microbalance (TEOM), filter dynamic measurement system (FDMS) and beta attenuation monitor (BAM) instruments, and the reasons for the drop in concentrations are complex. Previous studies indicated that the Fidas 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. As a result, and as also shown in the precursor to this study[9], 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.

This study is a review of PM_{10} and $PM_{2.5}$ data collected between January 2020 and June 2022 and builds on the results from the pilot project by strengthening the monitoring methodology to more closely follow BS EN 16450 – 'Automated measuring systems for the measurement of the concentration of particulate matter'. The primary change was the introduction of duplicate reference MPNS and Fidas instruments in line with BS EN 16450, providing a more robust dataset for identifying whether it would be appropriate to apply correction factors to PM_{10} and $PM_{2.5}$ Fidas data within the SAQD.

A key characteristic of the Scotland pollution climate is the relatively low PM concentrations. This is reflected in the number of days of PM_{10} and $PM_{2.5}$ above 28 and 17 $\mu g\ m^{-3}$, respectively, with a requirement for 20% of daily measurements to be greater than these limits – only 19 and 10 days or 5.1% and 3.2% of measurements were above these limits between 2020 and 2022.

Reviewing how the relationship between the reference method and Fidas varied with time and dataset used demonstrates that the relationship does vary. In particular, the range of results from regression analysis of PM_{10} and $PM_{2.5}$ measurements greater than the high thresholds of 18 and 30 $\mu g\ m^{-3}$, respectively, showed a wide variation. This, however, is likely to be a symptom of the low number of high thresholds days – the lower the number, the higher the uncertainty is in the intercept and slope results. The more data points collected above this threshold the better picture can be built up of how the analyser responds at these concentrations. Even though the 20% requirement was no met, however, this study has demonstrated the difficulty with achieving this. In general, this analysis confirmed that Fidas PM_{10} measurements did not achieve a measurement uncertainty of less than 25% without correction and that $PM_{2.5}$ measurements did pass the 25% criterion without correction.

Further analysis was carried out looking at how the Fidas PM measurement uncertainties might vary with applied slope and intercept. For PM_{10} this analysis verified that not applying a correction as is currently done would not optimise the measurement uncertainty. For $PM_{2.5}$, using the current correction by dividing by 1.06 still meets the 25% criterion but increases the measurement uncertainty from 10.7% to 16.5%, compared to not correcting the data. Using either 1.00 or 1.06 falls within the uncertainty band of the calculated slope of 1.033 ± 0.042 but the improvement in measurement uncertainty using uncorrected data indicates that no correction would be more appropriate.

Reviewing the results for the PM_{10} comparison using data from the duplicate instruments between 08/07/2021 to 29/06/2022 indicates that the Fidas does not pass the 25% criteria for measurement uncertainty (W_{CM}) when not corrected. When the data are corrected for both slope and intercept by adding 1.993 then dividing by 0.909 the Fidas meets the requirement for measurement uncertainty with a W_{CM} of 7.3% using all data. Simplifying

[9] Pilot Research Study to Investigate Particulate Matter Monitoring Techniques in Scotland, 19/08/2021: https://www.scottishairquality.scot/sites/default/files/publications/2021-09/Pilot_Research_Study_to_Investigate_PM_Monitorng_Techniques_in_Scotland_issue_1.pdf

the correction by dividing by 0.909 also meets this requirement with a W_{CM} of 11.0%. Table 15 summarises the PM_{10} results.

Table 15 Summary of measurement uncertainties of uncorrected and corrected PM_{10} Fidas data

Instrument	PM_{10} Measurement Uncertainty at 50 $\mu g\ m^{-3}$		
	No correction	Correction by dividing by adding 1.993 then 0.909	Correction by dividing by 0.909
Fidas 200	26.5%	7.3%	11.0%

Reviewing the results for the $PM_{2.5}$ comparison using data from the duplicate instruments between 08/07/2021 to 29/06/2022 (Table 16) indicates that the Fidas passes the 25% criterion with a W_{CM} of 10.7%. This is also the case for data corrected for intercept by adding 1.162 with a W_{CM} of 12.7% using all data. Using the current correction of gravimetric equivalence, 1.06, W_{CM} is 16.5%. These results indicate that no correction would be the most appropriate approach within the SAQD.

Table 16 Summary of measurement uncertainties of uncorrected and corrected $PM_{2.5}$ Fidas data

Instrument	$PM_{2.5}$ Measurement Uncertainty at 30 $\mu g\ m^{-3}$		
	No correction	Correction by adding 1.162	Correction by dividing by 1.06
Fidas 200	10.7%	12.7%	16.5%

Considering the results of this study, the following recommendations are made to the Scottish Government:

- As this study does not supersede the formal UK equivalence results, the corrections for gravimetric equivalence currently applied on the Scottish Air Quality Database and website[10] will remain unchanged.
- Fidas PM_{10} data collected within the SAQD should be corrected by dividing by 0.909, which should be applied to 2022 data onwards. A correction for slope only is recommended for simplicity and authorities will need to apply this correction to data downloaded from the Air Quality in Scotland website.
- Fidas $PM_{2.5}$ data collected within the SAQD should not be corrected. This will mean that authorities will need to apply a correction to data downloaded from the Air Quality in Scotland website by multiplying 1.06 applied to 2022 data onwards.
- For completeness, it is recommended that authorities report both the corrected, by applying the corrections defined above, and uncorrected results, as reported on the Air Quality in Scotland website within their reports.

[10] https://www.scottishairquality.scot/

APPENDICES

APPENDIX 1 CALCULATIONS USED

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:

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

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:

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

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

Where:

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

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

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

Where n is the number of paired measurements recorded.

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

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

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

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

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:

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

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:

$$|a| \le 2u_a \quad \text{(A12)}$$
$$|b-1| \le 2u_b \quad \text{(A13)}$$

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

UNCERTAINTY IN MEASUREMENTS

The between-reference method uncertainty, $u(bs, RM)$, is calculated as follows. If one reference sampler is used, then u_{RM} defaults to 0.67 µg m^{-3}.

$$u(bs, RM) = \frac{\sum_{i=1}^{n}\left(x_{i,1} - x_{i,2}\right)^2}{2n} \quad \text{(A14)}$$

Where:

$x_{i,1}, x_{i,2}$ are the results of parallel reference measurements for a single 24 hr period i.

n is the number of 24 hr measurement results.

A between-reference method uncertainty > 2,0 µg m^{-3} indicates that the performance of one or both instruments is unsuitable.

The between-candidate method uncertainty, $u(bs, CM)$, is calculated as follows:

$$u(bs, CM) = \frac{\sum_{i=1}^{n}\left(y_{i,1} - y_{i,2}\right)^2}{2n} \quad \text{(A15)}$$

Where:

$y_{i,1}, y_{i,2}$ are the results of parallel candidate measurements for a single 24 hr period i.

n is the number of 24 hr measurement results.

A between-candidate method uncertainty > 2.5 µg m^{-3} indicates that the performance of one or both instruments is unsuitable. Therefore, the dataset cannot be used as reference dataset.

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

$$RSS = \sum_{i=1}^{i=n} (y_i - a - bx_i)^2 \quad \text{(A16)}$$

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

$$\sigma = \left[\frac{1}{n-2}\sum_{i=1}^{i=n}(y_i - a - bx_i)^2 - {u_{RM}}^2\right]^{\frac{1}{2}} \quad \text{(A17)}$$

Where $u_{RM} = \frac{u(bs,RM)}{\sqrt{2}}$

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

$$[u(y_i)]^2 = \sigma^2 + [a + (b-1)z_i]^2 \quad \text{(A18)}$$

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:

$$w_h(y_i) = 100 \cdot \left(\frac{u(y_i)}{z_i}\right) \quad \text{(A19)}$$

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:

$$W(y_i) = k \cdot w_h(y_i) = 2w_h(y_i) \quad \text{(A20)}$$

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 $(y_{i,corr})$:

$$y_{i,corr} = y_i - a \quad \text{(A21)}$$

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

$$y_{i,corr} = c + dx_i \quad \text{(A22)}$$

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:

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

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 $(y_{i,corr})$:

$$y_{i,corr} = \frac{y_i}{b} \quad \text{(A24)}$$

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:

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

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 $(y_{i,corr})$:

$$y_{i,corr} = \frac{y_i - a}{b} \quad \text{(A26)}$$

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:

$$\left[u(y_{i,corr})\right]^2 = \sigma^2 + [c + ((d-1)z_i]^2 + z_i{}^2 u_b{}^2 + u_a{}^2 \quad \text{(A27)}$$

APPENDIX 2 REGRESSION RESULTS – ALL DATASETS

Table 17 Orthogonal regression results – PM_{10}

Glasgow Hope Street	Dataset	Orthogonal Regression – PM_{10}								LV of 50 µg m^{-3}	
		n	R^2	Slope (b)	±	ub	Intercept (a)	±	ua	W_{cm}	%>28
MPNS-1 vs Fidas-1 2020	Winter	74	0.94	0.776	±	0.023	0.158	±	0.398	44.50	6.8
	Summer	87	0.90	0.843	±	0.029	-0.924	±	0.406	35.39	1.1
	≥30	4	0.08	0.051	±	0.122	30.694	±	5.004	67.11	100.0
	<30	156	0.90	0.794	±	0.021	-0.245	±	0.301	42.30	0.0
	All data	161	0.93	0.800	±	0.017	-0.302	±	0.268	41.51	3.7
MPNS-1 vs Fidas-1 2021	Winter	59	0.62	1.114	±	0.089	-4.169	±	1.509	18.11	5.1
	Summer	88	0.94	0.849	±	0.022	-0.787	±	0.396	33.57	5.6
	≥30	6	0.01	-14.267	±	0.872	492.428	±	28.393	1088.39	100.0
	<30	141	0.70	0.950	±	0.044	-2.092	±	0.739	21.36	0.0
	All data	147	0.77	0.942	±	0.038	-1.988	±	0.658	22.31	5.4
MPNS-1 vs Fidas-1 2022	Winter	34	0.94	0.918	±	0.038	-0.656	±	0.886	21.43	23.5
	Summer	31	0.88	0.807	±	0.052	-0.034	±	0.869	39.32	6.5
	≥30	8	0.75	0.820	±	0.172	2.719	±	6.674	27.56	100.0
	<30	57	0.82	0.903	±	0.052	-0.989	±	0.844	24.82	0.0
	All data	65	0.93	0.915	±	0.030	-1.138	±	0.613	23.27	15.4
MPNS-1 vs Fidas-1 All data	Winter	167	0.85	0.910	±	0.028	-1.310	±	0.516	25.88	9.6
	Summer	206	0.93	0.847	±	0.016	-0.841	±	0.262	34.18	3.9
	≥30	18	0.54	0.711	±	0.132	5.649	±	4.934	37.47	100.0
	<30	354	0.79	0.890	±	0.022	-1.269	±	0.341	28.18	0.0
	All data	373	0.88	0.892	±	0.016	-1.276	±	0.282	28.09	6.4
MPNS-2 vs Fidas-1 2021	Winter	23	0.60	0.558	±	0.087	3.264	±	1.216	75.52	0.0
	Summer	43	0.88	0.960	±	0.053	-2.539	±	0.808	18.76	0.0
	≥30	0	-	-	±	-	-	±	-	-	0.00
	<30	66	0.79	0.865	±	0.050	-1.048	±	0.737	31.70	0.0
	All data	66	0.79	0.865	±	0.050	-1.048	±	0.737	31.70	0.0
MPNS-2 vs Fidas-1 2022	Winter	40	0.97	0.939	±	0.024	-0.388	±	0.618	15.82	25.0
	Summer	46	0.92	0.898	±	0.039	-0.682	±	0.599	23.55	2.2
	≥30	9	0.98	0.963	±	0.047	-1.406	±	2.089	14.28	100.0
	<30	77	0.87	0.916	±	0.038	-0.587	±	0.598	20.08	0.0
	All data	86	0.97	0.950	±	0.018	-1.065	±	0.372	15.62	12.8
MPNS-2 vs Fidas-1 All data	Winter	63	0.96	0.953	±	0.024	-1.198	±	0.521	16.41	15.9
	Summer	89	0.89	0.927	±	0.034	-1.568	±	0.513	21.37	1.1
	≥30	9	0.98	0.963	±	0.047	-1.406	±	2.089	14.28	100.0
	<30	143	0.83	0.920	±	0.032	-1.193	±	0.488	21.75	0.0
	All data	152	0.95	0.961	±	0.017	-1.760	±	0.319	16.32	7.1
	Winter	22	0.47	0.448	±	0.092	3.386	±	1.268	96.95	5.1

Glasgow Hope Street	Dataset	Orthogonal Regression – PM_{10}								LV of 50 µg m^{-3}	
		n	R^2	Slope (b)	±	ub	Intercept (a)	±	ua	W_{cm}	%>28
MPNS-1 vs Fidas-2 2021	Summer	61	0.88	0.787	±	0.036	-1.204	±	0.557	47.49	5.6
	≥30	18	0.54	0.711	±	0.132	5.649	±	4.934	37.47	100.0
	<30	83	0.80	0.729	±	0.038	-0.342	±	0.568	55.68	0.0
	All data	83	0.80	0.729	±	0.038	-0.342	±	0.568	55.68	5.4
MPNS-1 vs Fidas-2 2022	Winter	34	0.94	0.842	±	0.037	-0.566	±	0.851	35.20	23.5
	Summer	31	0.93	0.715	±	0.035	0.221	±	0.591	56.33	6.5
	≥30	8	0.73	0.790	±	0.172	0.739	±	6.659	40.60	100.0
	<30	57	0.83	0.844	±	0.047	-1.156	±	0.765	36.68	0.0
	All data	65	0.93	0.837	±	0.027	-1.056	±	0.556	37.65	15.4
MPNS-1 vs Fidas-2 All data	Winter	56	0.92	0.852	±	0.033	-1.254	±	0.667	35.86	9.6
	Summer	92	0.90	0.752	±	0.025	-0.576	±	0.401	51.95	3.9
	≥30	8	0.73	0.790	±	0.172	0.739	±	6.659	40.60	100.0
	<30	140	0.81	0.806	±	0.031	-1.104	±	0.479	43.67	0.0
	All data	148	0.91	0.836	±	0.021	-1.522	±	0.368	39.43	6.4
MPNS-2 vs Fidas-2 2021	Winter	23	0.62	0.522	±	0.080	2.553	±	1.113	85.42	0.0
	Summer	43	0.85	0.854	±	0.052	-2.222	±	0.786	38.31	0.0
	≥30	0	-	-	±	-	-	±	-	-	0.0
	<30	66	0.79	0.772	±	0.046	-0.944	±	0.680	49.62	0.0
	All data	66	0.79	0.772	±	0.046	-0.944	±	0.680	49.62	0.0
MPNS-2 vs Fidas-2 2022	Winter	40	0.97	0.852	±	0.023	-0.203	±	0.593	31.29	25.0
	Summer	46	0.95	0.797	±	0.028	-0.348	±	0.433	42.03	2.2
	≥30	9	0.98	0.890	±	0.051	-2.031	±	2.283	30.79	100.0
	<30	77	0.88	0.841	±	0.034	-0.584	±	0.540	34.53	0.0
	All data	86	0.97	0.863	±	0.016	-0.898	±	0.338	31.58	12.8
MPNS-2 vs Fidas-2 All data	Winter	63	0.96	0.870	±	0.023	-1.164	±	0.498	31.56	15.9
	Summer	89	0.89	0.825	±	0.030	-1.258	±	0.459	40.21	1.1
	≥30	9	0.98	0.890	±	0.051	-2.031	±	2.283	30.79	100.0
	<30	143	0.83	0.843	±	0.030	-1.235	±	0.460	36.79	0.0
	All data	152	0.95	0.875	±	0.016	-1.686	±	0.300	32.29	7.1

Table 18 Orthogonal regression results – $PM_{2.5}$

Glasgow Hope Street	Dataset	Orthogonal Regression – $PM_{2.5}$								LV of 30 µg m^{-3}	
		n	R^2	Slope (b)	±	ub	Intercept (a)	±	ua	W_{cm}	%>17
MPNS-1 vs Fidas-1 2020	winter	52	0.86	0.712	±	0.038	0.691	±	0.403	53.71	11.5
	summer	83	0.69	0.931	±	0.058	-1.388	±	0.534	26.05	1.2
	≥18	5	0.84	1.450	±	0.320	-15.335	±	6.918	15.17	100.0
	<18	130	0.69	0.848	±	0.043	-0.568	±	0.387	35.94	0.0
	all data	135	0.77	0.804	±	0.034	-0.254	±	0.337	42.20	5.2
MPNS-1 vs Fidas-1 2021	winter	87	0.78	0.940	±	0.048	-0.340	±	0.380	17.22	1.1
	summer	58	0.81	1.071	±	0.062	-2.720	±	0.758	14.36	13.8
	≥18	4	0.99	2.026	±	0.132	-25.034	±	3.050	38.31	100.0
	<18	141	0.75	0.906	±	0.039	-0.426	±	0.359	24.40	0.0
	all data	145	0.82	0.950	±	0.034	-0.782	±	0.338	19.23	6.1
MPNS-1 vs Fidas-1 2022	winter	27	0.82	0.728	±	0.064	2.078	±	0.615	41.51	3.7
	summer	38	0.82	0.979	±	0.070	-0.684	±	0.599	12.48	0.0
	≥18	4	0.99	2.026	±	0.132	-25.034	±	3.050	38.31	100.0
	<18	64	0.75	0.867	±	0.056	0.496	±	0.486	25.24	0.0
	all data	65	0.80	0.860	±	0.050	0.547	±	0.447	26.20	1.5
MPNS-1 vs Fidas-1 All data	winter	166	0.80	0.793	±	0.029	0.599	±	0.261	38.69	4.8
	summer	179	0.78	0.990	±	0.035	-1.631	±	0.356	17.77	5.0
	≥18	10	0.85	2.182	±	0.284	-29.943	±	6.345	39.32	100.0
	<18	335	0.71	0.883	±	0.026	-0.365	±	0.239	28.11	0.0
	all data	345	0.78	0.884	±	0.023	-0.390	±	0.220	28.32	4.9
MPNS-2 vs Fidas-1 2021	winter	32	0.82	1.232	±	0.095	-1.529	±	0.793	38.19	2.9
	summer	22	0.89	1.280	±	0.092	-3.974	±	1.130	31.80	9.1
	≥18	3	0.76	2.361	±	1.062	-26.568	±	22.655	99.22	100.0
	<18	51	0.74	1.077	±	0.078	-0.959	±	0.711	14.58	0.0
	all data	54	0.85	1.169	±	0.062	-1.719	±	0.632	25.71	5.4
MPNS-2 vs Fidas-1 2022	winter	10	0.58	0.946	±	0.219	0.700	±	1.535	11.28	0.0
	summer	39	0.69	1.087	±	0.098	-1.343	±	0.816	14.56	0.0
	≥18	-	-	-	±	-	-	±	-	-	0.0
	<18	49	0.67	1.057	±	0.088	-0.890	±	0.713	12.73	0.0
	all data	49	0.67	1.057	±	0.088	-0.890	±	0.713	12.73	0.0
MPNS-2 vs Fidas-1 All data	winter	42	0.79	1.204	±	0.085	-1.240	±	0.687	34.37	2.3
	summer	61	0.83	1.156	±	0.061	-2.116	±	0.607	20.86	3.3
	≥18	3	0.76	2.361	±	1.062	-26.568	±	22.655	99.22	100.0
	<18	100	0.71	1.072	±	0.057	-0.957	±	0.495	13.89	0.0
	all data	103	0.81	1.143	±	0.049	-1.510	±	0.456	22.23	2.9
MPNS-1 vs Fidas-2 2021	winter	41	0.80	1.016	±	0.072	-1.636	±	0.660	13.21	1.1
	summer	37	0.88	1.017	±	0.060	-2.870	±	0.792	18.89	13.8
	≥18	3	0.93	2.088	±	0.525	-28.532	±	12.668	30.22	100.0
	<18	75	0.78	0.927	±	0.051	-1.327	±	0.530	25.60	0.0
	all data	78	0.85	0.963	±	0.042	-1.668	±	0.478	21.47	6.1

Glasgow Hope Street	Dataset	Orthogonal Regression – $PM_{2.5}$								LV of 30 µg m^{-3}	
		n	R^2	Slope (b)	±	ub	Intercept (a)	±	ua	W_{cm}	%>17
MPNS-1 vs Fidas-2 2022	winter	27	0.81	0.674	±	0.061	1.976	±	0.585	52.67	3.7
	summer	38	0.81	0.887	±	0.066	-0.308	±	0.565	25.95	0.0
	≥18	3	0.93	2.088	±	0.525	-28.532	±	12.668	30.22	100.0
	<18	64	0.74	0.785	±	0.052	0.719	±	0.452	39.11	0.0
	all data	65	0.79	0.783	±	0.046	0.737	±	0.416	39.47	1.5
MPNS-1 vs Fidas-2 All data	winter	68	0.77	0.866	±	0.052	-0.089	±	0.483	29.44	4.8
	summer	75	0.86	0.906	±	0.040	-0.974	±	0.445	27.10	5.0
	≥18	4	0.94	2.105	±	0.353	-28.983	±	8.391	29.08	100.0
	<18	139	0.76	0.846	±	0.036	-0.177	±	0.350	33.55	0.0
	all data	143	0.82	0.880	±	0.031	-0.474	±	0.324	28.98	4.9
MPNS-2 vs Fidas-2 2021	winter	32	0.81	1.201	±	0.094	-1.860	±	0.788	30.17	2.9
	summer	22	0.89	1.149	±	0.084	-3.423	±	1.029	12.59	9.1
	≥18	3	0.61	2.457	±	1.295	-30.501	±	27.640	95.69	100.0
	<18	51	0.72	1.001	±	0.075	-0.970	±	0.683	12.56	0.0
	all data	54	0.84	1.083	±	0.060	-1.650	±	0.613	13.31	5.4
MPNS-2 vs Fidas-2 2022	winter	10	0.58	0.875	±	0.208	0.733	±	1.458	21.90	0.0
	summer	39	0.69	0.978	±	0.090	-0.865	±	0.752	14.83	0.0
	≥18	-	-	-	±	-	-	±	-	-	0.0
	<18	49	0.66	0.952	±	0.081	-0.486	±	0.657	16.50	0.0
	all data	49	0.66	0.952	±	0.081	-0.486	±	0.657	16.50	0.0
MPNS-2 vs Fidas-2 All data	winter	42	0.79	1.168	±	0.084	-1.513	±	0.680	25.98	2.3
	summer	61	0.82	1.026	±	0.056	-1.518	±	0.558	11.98	3.3
	≥18	3	0.61	2.457	±	1.295	-30.501	±	27.640	95.69	100.0
	<18	100	0.70	0.977	±	0.054	-0.723	±	0.464	14.08	0.0
	all data	103	0.80	1.045	±	0.047	-1.251	±	0.432	11.41	2.9

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