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Discussions

Air pollution measurement errors: Is your data fit for purpose?

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Abstract. When making measurements of air quality, having a reliable estimate of the measurement uncertainty is key to assessing the information content that an instrument is capable of providing, and thus its usefulness in a particular application. This is especially important given the widespread emergence of Low Cost Sensors (LCS) to measure air quality. To do this, end users need to clearly identify the data requirements a priori and design quantifiable success criteria by which to judge the data. All measurements suffer from errors, with the degree to which these impact the accuracy of the final data often determined by our ability to identify and correct for them. The advent of LCS has provided a challenge in that many error sources show high spatial and temporal variability, making laboratory derived corrections difficult. Characterizing LCS performance thus currently depends primarily on colocation studies with reference instruments, which are very expensive and do not offer a definitive solution but rather a glimpse of LCS performance in specific conditions over a limited period of time. Despite the limitations, colocation studies do provide useful information on measurement device error structure, but the results are non-trivial to interpret and often difficult to extrapolate to future device performance. A problem that obscures much of the information content of these colocation performance assessments is the exacerbated use of global performance metrics (R^2, RMSE, MAE, etc.). Colocation studies are complex and time-consuming, and it is easy to fall into the temptation to only use these metrics when trying to define the most appropriate sensor technology to subsequently use. But the use of these metrics can be limited, and even misleading, restricting our understanding of the error structure and therefore the measurements' information content. In this work, the nature of common air pollution measurement errors is investigated, and the implications these have on traditional metrics and other empirical, potentially more insightful, approaches to assess measurement performance. With this insight we demonstrate the impact these errors can have on measurements, using a selection of LCS deployed alongside reference measurements as part of the QUANT project, and discuss the implications this has on device end-use.

Keywords: air quality measurements, errors, uncertainty, information content, low-cost sensors.

1. Introduction

The measurement of air pollutants is central to our ability to both devise and assess the effectiveness of policies to improve air quality and reduce human exposure (Molina & Molina, 2004). The emergence of low-cost sensor (LCS) based technologies means a growing number of measurement devices are now available for this purpose (Morawska et al., 2018), ranging from small low-cost devices that can be carried on an individual's person all the way through to large, expensive reference and research-grade instrumentation. A key question that needs to be asked when choosing a particular measurement technology is whether the data provided is fit for purpose (Andrewes et al., 2021; Lewis & Edwards, 2016). In order to answer this, the user must first clearly define the question that is to be asked of the data, and thus the information required. For example, a measurement to characterize "rush hour" concentrations, or to determine if the concentration of a pollutant exceeded an 8 h average legal threshold value at a particular location would demand a very different set of data requirements than a measurement to determine if a change in policy had modified the average pollutant concentration trend in a neighbourhood. Considerations such as measurement time resolution and ability to capture spatial variability would be important for such examples (Feinberg et al., 2019). The measurement uncertainty is also of critical consideration, as this ultimately determines the information content of the data, and hence how it can be used (Tian et al., 2016).

All measurements have an associated uncertainty, and even in highly controlled laboratory assessments, the true value is not known, with any measurement error defined relative to our best estimate of the range of possible true values. However, quantifying and representing error and uncertainty is a challenge for a wide range of analytical fields, and often what these concepts represent is not the same to all practitioners. This results in a spectrum of definitions that take into account the way truth, error, and uncertainty are conceived (Cross et al., 2017; Grégis, 2019; Kirkham et al., 2018; Mari et al., 2021). For atmospheric measurements assessing uncertainty is complex and non-trivial. Firstly, given the "true" value can never be known, an agreed reference is needed. Secondly, the constantly changing atmospheric composition means that repeat measurements cannot be made and the traditional methods for determining the random uncertainty are not applicable. And finally, a major challenge arises from the multiple sources of error both internal and external to the sensor that can affect a measurement. Signal responses from a non-target chemical or physical parameter or electromagnetic interference are examples of an almost limitless number of potential sources of measurement error. In this work, we will follow the definitions given by the International Vocabulary of Metrology (JCGM, 2012) for measurement error ("measured quantity value minus a reference quantity value") and for measurement uncertainty ("non-negative parameter characterizing the dispersion of the quantity values being attributed to a measurand, based on the information used").

The difficulties in defining and quantifying uncertainty across the full range of end-use applications of a measurement device, means that often the quoted measurement uncertainty is not applicable, or in some cases not provided or provided in an ambiguous manner. This makes assessing the applicability of a measurement device to a particular task difficult for end-users. In this work, we investigate the nature of common air pollution measurement errors, and the implications these have on traditional goodness-of-fit metrics and other, potentially more insightful approaches to assess measurement uncertainty. We then use this insight to demonstrate the impact these errors can have on measurements, using a selection of LCS deployed alongside reference measurements as part of the UK Clean Air program funded QUANT (Quantification of Utility of Atmospheric Network Technologies) project, a 2 year colocation study of 26 commercial LCS devices (56 gases measurements and 56 PM measurements) at multiple urban, background and roadside, locations in the UK. After analysing some of the real-life uncertainty characteristics we discuss the implications this has on data use.

2. Error characterization

When characterizing measurement error, in the absence of evidence to the contrary it is often assumed a linear additive model. Once the analytical form of the model is defined, its parameters aim to capture the error characteristics, and in the case of linear models (Eq. (1)), these are typically separated into three types (Tian et al., 2016): (i) proportional bias or scale error (b_1), (ii) constant bias or displacement error (b_0) and (iii) random error (ε) (Tian et al., 2016). Any measurement (y_i, e.g from the LCS) can therefore be thought of as a combination of the reference value (x_i) and the three error types, such that:

$$y_i = b_1 x_i + b_0 + \varepsilon \qquad (1)$$

As the simplest approximation, this linear relationship for the error characteristics is often used to correct for observed deviations between measurements and the agreed reference. It is worth to note, however, that this equation assumes time-independent error contributions and that the three error components are not correlated, which is often not the case on both counts (e.g. responses to non-target compounds). The parameter values determined for Eq. (1) are also generally only applicable for individual instruments, potentially in specific environments, unless the transferability of these parameters between devices has been explicitly demonstrated.

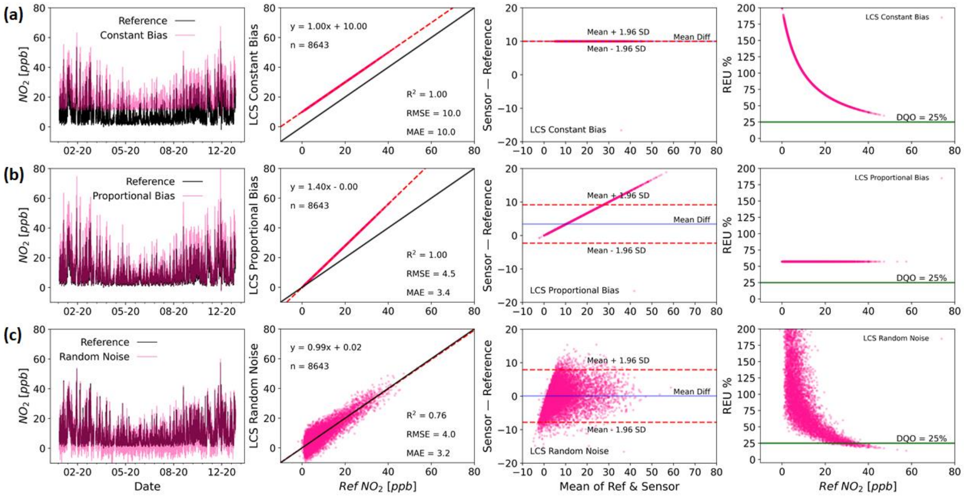

Figure 1. Time series (left panels), regression plots (middle-left panels), Bland-Altman plots (middle-right panels) and REU (right panels, DQO for NO_2 = 25%) for arbitrary examples of pure constant bias (Slope = 1, Intercept = 1, SD_ε = 0; a-panels), pure proportional bias (Slope = 1.4, Intercept = 0, SD_ε = 0; b-panels) and pure random noise (Slope = 1, Intercept = 0, SD_ε = 4; c-panels) simulated errors.

Figure 1 shows examples of how pure constant bias (a-panels), pure proportional bias (b-panels), and pure random noise (c-panels) would look like. In each of these ideal cases, the error plots enable the practitioner to view the error characteristics in slightly different ways, allowing the impacts of the observed measurement uncertainty to be placed into the context of the data requirements. In this work, we will refer to them as "error types" (in contrast to "error sources"), which is the way they are distilled by the linear error model.

2.1 Performance indices, error structure and uncertainty

A major challenge faced by end-users of measurement devices characterised using colocation studies is the non-trivial question of how the comparisons themselves are performed and how the data is communicated. Often single value performance metrics, such as the coefficient of determination (R^2) or root mean squared error (RMSE), are calculated between the assessed method (e.g. LCS) and an agreed reference, and the user is expected to infer an expected device performance or uncertainty for a measurement in their application (Cross et al., 2017; Duvall et al., 2016; Malings et al., 2019). These metrics contain useful information about the measurement, but they are unable to fully describe the error characteristics, in part because they reduce the error down to a single value (Tian et al., 2016). Unfortunately, the widespread use of a small number of metrics as the sole method to assess measurement uncertainty, without a thorough consideration of the nature of the measurement errors, means measurement devices are often chosen that are unable to provide data that is fit for purpose. In addition, unconscious about potential flaws, users (e.g. researchers) could communicate findings or guide decision making based on results that may not justify the conclusions drawn from the data. Figure 2 shows three simulated measurements compared with the true values. Despite the measurements having identical R^2 and RMSE values, the time series and regression plots show that the error characteristics are significantly different, and would impact how the data from such a device could viably be used.

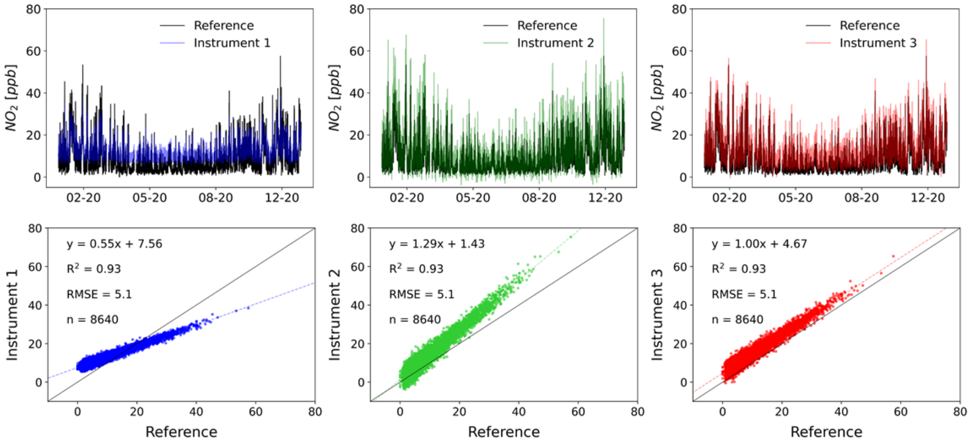

Figure 2. Time series (upper panels) and regression plots (lower panels) for three hypothetical instruments and a reference. The most used metrics for evaluating the performance of LCS (R^2 and RMSE) are identical for the systems shown, even when the errors have very different characteristics (time res 1 h).

There are multiple performance metrics that can be used for the assessment of measurement errors and uncertainty. Tian et al (2016) present an excellent summary of some of the major pitfalls of performance metrics and promote an approach of error modelling as a more reliable method of uncertainty quantification. These modelling approaches, however, rely on the assumption of statistical stationarity, whereby the statistical properties of the error are constant in the temporal and spatial domains. The presence of unknown or poorly characterised sources of error, for example, due to interferences from other atmospheric constituents or drifts in sensor behaviour, makes this assumption difficult to satisfy, especially when the dependencies of these errors show high spatial and temporal variability. Thus, if field colocation studies are the primary method for performance assessment, as is the case for LCS, only through a detailed assessment of the measurement errors across a wide range of conditions and timescales can the uncertainty of the measurement be realistically estimated.

2.2 Dealing with errors: established techniques vs Low-Cost Sensors

Different approaches are available to the user to minimise the impact of errors, generally by making corrections to the sensor data. For example, in the case of many atmospheric gas analysers, if the error is dominated by a proportional bias, a multi-point calibration can be performed using standard additions of the target gas. Displacement errors can be quantified, and then corrected for, by sampling a gas stream that contains zero target gas. And random errors can be reduced by applying a smoothing filter (e.g moving average filter, time-averaging the data, etc.), at the cost of losing some information (Brown et al., 2008). These approaches work well for simple error sources that, ideally, do not change significantly over timescales from days to months. Unfortunately, more complex error sources can manifest in such a way that they contribute across all three error types, and also vary temporally and spatially. For example, an interference from another gas-phase compound could in part manifest itself as a displacement error, based on the instrument response to its background value, and in part as a proportional bias if its concentration correlates with the target compounds, with any short-term deviations from perfect correlation contributing to the random error component. In this case, time-averaging combined with periodic calibrations and zeros would not necessarily minimise the error, and the user would need to employ different tactics. One option would be to independently measure the interferent concentration, albeit with associated uncertainty, and then use this to derive a correction. This is feasible if a simple and cost-effective method exists for quantifying the interferent and its influence on the result is understood, but can make it very difficult to separate out error sources, and can become increasingly complex if this measurement also suffers from other interferences.

For many measurement devices, in particular for LCS based instruments, a major challenge is that the sources and nature of all the errors are unknown or difficult to quantify across all possible end-use applications, meaning estimates of measurement uncertainty are difficult. In the case of most established research and reference-grade measurement techniques, comprehensive laboratory and field experiments have been used to explore the nature of the measurement errors (Gerboles et al., 2003; Zucco et al., 2003). Calibrations have then been developed, where traceable standards are sampled and measurement bias, both constant and proportional, can be corrected for. Interferences from variables such as temperature, humidity, or other gases, have also been identified and then either a solution engineered to minimise their effect or robust data corrections derived. Unfortunately, these approaches have been shown not to perform well in the assessment of LCS measurement errors, due to the presence of multiple, potentially unknown, sensor interferences from other atmospheric constituents (Thompson & Ellison, 2005). These significant sensitivities to constituents such as water vapour and other gases mean laboratory-based calibrations of LCS become exceedingly complex, and expensive, as they attempt to simulate the true atmospheric complexity, often resulting in observed errors being very different to real-world sampling (Rai et al., 2017; Williams, 2020). This has resulted in colocation calibration becoming the accepted method for characterizing LCS measurement uncertainties (De Vito et al., 2020; Masson et al., 2015; Mead et al., 2013; Popoola et al., 2016; Sun et al., 2017), where sensor devices are run alongside traditional reference measurement systems for a period of time, and statistical corrections derived to minimise the error between the two. As the true value of a pollutant concentration cannot be known, this colocation approach assumes all the error is in the low-cost measurement. Although this assumption may often be approximately valid (i.e. reference error variance << LCS error variance), no measurement is absent of uncertainty and this can be transferred from one measurement to another, obscuring attempts to identify its sources and characteristics. A further consideration when the fast time-response aspect of LCS data is important, is that reference measurement uncertainties are generally characterised at significantly lower reported measurement frequencies (typically 1 hr). This means that a high time-resolution (e.g. 1 min) reference uncertainty must be characterized in order to accurately estimate the LCS uncertainty (requiring specific experiments and additional costs). If a lower time-resolution reference data is used as a proxy, then the natural variability timescales of the target compound should be known and any impact of this on the reported uncertainty caveated.

Another challenge with this approach is that, unlike targeted laboratory studies, real-world colocation studies at a single location, and for a limited time period, are not able to expose the measurement devices to the full range of potential sampling conditions. As many error sources are variable, both spatially and temporally, using data generated under a limited set of conditions to predict the uncertainty on future measurements is risky. Deploying a statistical model makes the tacit assumption that all factors affecting the target variable are captured by the model (and the data set used to build the model). This is very often an unrealistic demand, and in the complex multifaceted system that is atmospheric chemistry, this is extremely unlikely to be tenable, resulting in a clear potential for overfitting to the training dataset. Ultimately, however, these colocation comparisons with instruments with a well-quantified uncertainty need to be able to communicate a usable estimate of the information content of the data to end-users, so that devices can be chosen that are fit for a particular measurement purpose.

3. Methods

In this work, we explore measurement errors, and their impacts, using the most common single value metrics (R^2, RMSE and MAE), along with two additional widely used approaches to visualize the error across a dataset: the Bland-Altman plots (B-A) and Relative Expanded Uncertainty (REU).

The performance metrics provide a single value irrespective of the size of the dataset, and might appear convenient for users when comparing across devices or datasets, but can encourage over-reliance on the metric, often at the expense of looking at the data in more detail or bringing an awareness of the likely physical processes driving the error sources. On the other hand, the B-A and the REU plots are more laborious techniques and the interpretation can be challenging for non-experts, but they provide additional insights into the nature of the errors, not attainable by one or more combined performance metrics.

In order to understand how the different tools used here show different characteristics of the error structure, some errors commonly found in LCS are examined through simulation studies. Subsequently, two real world case studies are presented: (i) LCS duplicates for NO_2 and $PM_{2.5}$ belonging to the QUANT project located in two sites -the Manchester Natural Environment Research Council (NERC) measurement Supersite, and the York Fishergate Automatic Urban and Rural Network (AURN) roadside site- and (ii) a set of duplicate reference instruments (only at Manchester Supersite). Table S1 shows the research grade instrumentation used for this study.

3.1 Visualization tools

An ideal performance metric should be able to deliver not only a performance index but also an idea of the uncertainty distribution (Chai & Draxler, 2014). This is difficult to deliver through a simple numerical value, and easy to interpret visualisations of the data are often much more useful for conveying multiple aspects of data performance. Figure 2 shows the two most common data visualisation tools, the time-series plot and the regression plot. In the time series plot the instrument under analysis and the agreed reference are plotted together as a function of time. This allows a user to visually assess tendencies of over or under prediction, differences in the base line or other issues, but can be readily over interpreted and does not allow for easy quantification of the observed errors. In the regression plot the data from the instrument under analysis is plotted against the agreed reference data. This allows for the correlation between the two methods to be more readily interpreted, in particular any deviations from linearity, but gives little detail on the nature of the errors themselves.

In contrast to the regression plot, Bland-Altman (B-A) plots essentially display the difference between measurements, enabling more information on the nature of the error to be communicated. B-A plots (Altman & Bland, 1983) are extensively used in analytical chemistry and biomedicine to evaluate the differences between two measurement techniques (Doğan, 2018). The B-A is a scatter plot, in which the abscissa represents the average of these measures (e.g LCS and a reference measurement), acknowledging that the true value is unknown and that both measurements have errors, and the ordinate shows the difference between the two paired measurements. In the case where all the error is assumed to be in one of the measurements, e.g. comparing a LCS to a reference grade measurement, there is an argument that the B-A abscissa could be the agreed reference value instead of the average of two measurements. However, in this work we use the average of the two values as per the traditional B-A analysis. To illustrate the B-A interpretation, from the error model (Eq. (1)) we can derive the following expression:

$$y_i - x_i \; = \; x_i \, (\, b_1 - 1) \; + \; b_0 \; + \varepsilon \qquad (2)$$

From Eq. (2) it can be seen that if $b_1 \neq 1$ or if the error term (ε) variance is non-constant (e.g. heteroscedasticity) the difference will not be normally distributed. The B-A plot (with x_i as the reference instrument results) allows a quick visual assessment of the error distribution without the need to calculate the model parameters. In the case the differences are normally distributed, the so-called "agreement interval" (usually defined as ± 2σ around the mean) will hold 95% of the data points. Even though the estimated limits of agreement will be biased if the differences are not normally distributed, it can still be a valuable indicator of agreement between the two measurements.

If the ultimate goal of studying measurement errors is to diagnose the measurement uncertainty in a particular target measurement range, then visualising the uncertainty in pollutant concentration space can be very informative. The REU (GDE, 2010) provides a relative measure of the uncertainty interval about the measurement within which the true value can be confidently asserted to lie. The abscissa in an REU plot represents the agreed reference pollutant concentration, whose error is taken into account, something not considered by the other metrics or visualisations discussed. The REU is regularly used to assess measurement compliance with the Data Quality Objective (DQO) of the European Air Quality Directive 2008/50/EC, and is mandatory for the demonstration of equivalence of methods other than the EU reference methods. For LCS the REU is widely used as a performance indicator (Bagkis et al., 2021; Bigi et al., 2018; Castell et al., 2017; Cordero et al., 2018; Spinelle et al., 2015). However, the evaluation of this metric is perceived as arduous and cumbersome and it is not included in the majority of sensor studies (Karagulian et al., 2019). There is now a new published European Technical Specification (TS) for evaluating the LCS performance for gaseous pollutants (CEN/TS 17660-1:2021). It categorizes the devices in 3 classes according to the DQO (Class 1 for "indicative measurements", Class 2 for "objective estimations", and Class 3 for non-regulatory purposes, e.g. research, education, citizen science, etc.).

In the following sections, we use these established methods for assessing measurement uncertainty, alongside simple time series and regression plots, to explore different error sources and their implications for air pollution measurements.

4. Case studies

4.1 Simulated instruments

In order to investigate the impact of different origins of measurement error on measurement performance, a set of simulated datasets have been created. These data are derived using real-world reference data as the true values, with the subsequent addition of errors of different origins to generate the simulated measurement data. Error origins were chosen for which examples have been described in the LCS literature. Performance metrics along with visualization methods are then used to assess measurement performance.

As the complexity of the error increases, the impact of the assumption of statistical stationarity can become more difficult to satisfy, with the magnitude of the errors becoming less uniform across the observed concentration, and hence spatial, or time domains. Figure 3 shows examples of modelled sources of errors on NO_2 measurements: temperature interference (correction model taken from (Popoola et al., 2016), a-panels), a non-target gas (ozone) interference (correction model taken from (Peters et al., 2021), b-panels) and thermal electrical noise (white noise, c-panels).

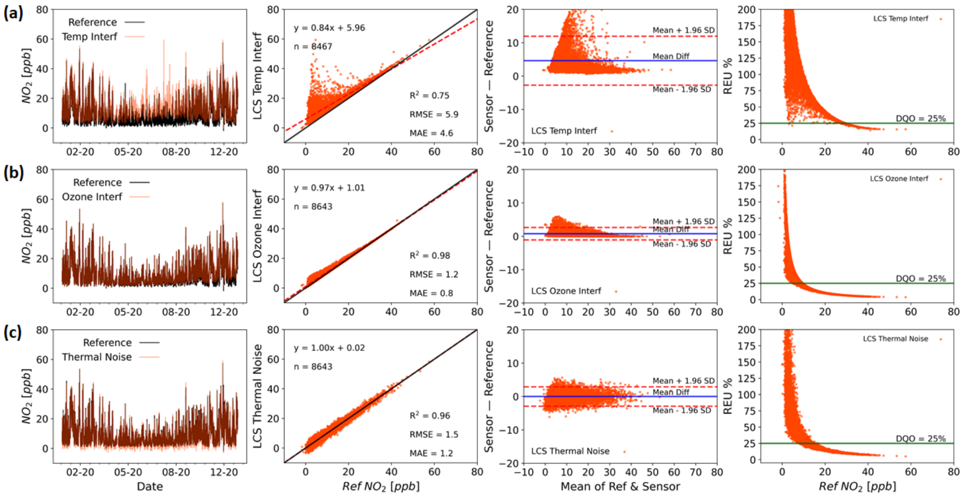

Figure 3. Time series (left panels), regression plots (middle-left panels, including R^2, RMSE & MAE), Bland-Altman plots (middle-right panels) and REU (right panels, DQO for NO_2 = 25%) for temperature (a-panels), ozone (b- panels) and thermal electrical noise (c-panels) modelled interferences on NO_2 measurements (time res 1 h).

The above simulations show examples of how individual sources of error can impact measurement performance. Figure S1 shows some more examples, this time for different drift effects (baseline drift, temperature interference drift and instrument sensitivity drift). This set of error origins is not exhaustive, with countless others potentially impacting the measurement, such as those coming from (i) hardware (sensor-production variability, sampling, thermal effects due to materials expansion, drift due to ageing, RTC lag, Analog-to-Digital conversion, electromagnetic interference, etc.), (ii) software (signal sampling frequency, signal-to-concentration conversion, concept drift, etc.), (iii) sensor technology/measurement method (selectivity, sensitivity, environmental interferences, etc.) and (iv) local effects (spatio-temporal variation of concentrations, turbulence, sampling issues etc.).

Each error source impacts the uncertainty of the measurement, which in turn impacts its ability to provide useful information for a particular task. For example, the form of the temperature interference shown in Fig. 3 (a-panels) results in the largest errors being seen at the lower NO_2 values. This is because NO_2 concentrations are generally lowest during the day, due to photolytic loss when temperatures are highest. Thus this device would be better suited to an end-user intending to assess daily peak NO_2 concentration compared with the daytime hourly exposure values, providing the environment the device was deployed in showed a similar relationship between temperature and true NO_2 as that used here. The O_3 interference shown in Fig. 3 (b-panels) is similar, due again to a general anti-correlation observed between ambient O_3 and NO_2 concentrations. This type of interference can often be interpreted incorrectly as a proportional bias, and a slope correction applied to the data. However, this type of correction will ultimately fail as O_3 concentrations are dependent on a range of factors, such as hydrocarbon concentrations and solar radiation, and as these change the O_3 concentration relative to the NO_2 concentration will change. To further complicate matters, multiple error sources can act simultaneously, meaning that the majority of measurements will contain multiple sources of error. Figure 4 shows a simple linear combination of the modelled errors shown in Fig 3, and the impact this has on the performance metrics.

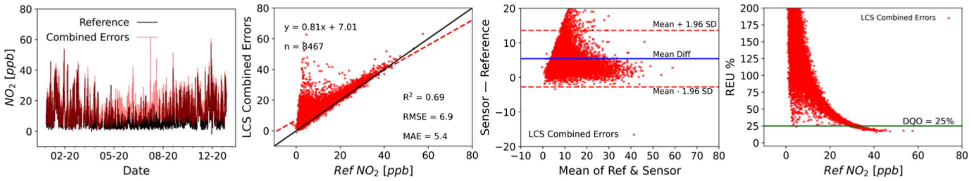

Figure 4. Time series (left panel), regression plot (middle-left panel, including R^2, RMSE & MAE), Bland-Altman plot (middle-right panel) and REU (right panel, DQO for NO$_2$ = 25%) for a linear combination of temperature, ozone and thermal electrical noise modelled interferences (time res 1 h).

As the simulations show, the nature of the errors determine the observed effect on the measurement performance. In an ideal situation, like those shown in figures 3 and 4, the error sources would be well characterised, allowing the error to be modelled and approaches such as calibrations (for bias) and smoothing (for random errors) employed to minimise the total uncertainty. Unfortunately, in scenarios where sources of error and their characteristics are not known, modelling the error becomes more difficult and a more empirical approach to assessing the measurement performance and uncertainty may be required. The growing use of LCS represents a particular challenge in this regard. The susceptibility of LCS to multiple, often unknown or poorly characterised, error sources means that in order to determine if a particular LCS is able to provide data with the required level of uncertainty for a given task, a relevant uncertainty assessment is required. The following section explores the uncertainty characteristics of several LCS, with unknown error sources, deployed alongside reference instrumentation in UK urban environments as part of the QUANT study.

4.2 Real-world instruments

The difficulty in generating representative laboratory error characterisation data means for many measurement devices the error sources are essentially unknown. This, combined with the use of imperfect algorithms that are not available to the end-user (i.e. "black-box" models) to minimise errors, means that, colocation data is often the best option available to end-users in order to assess the applicability of a measurement method for their desired purpose. This is particularly the case for LCS air pollution measurement devices. In this section, we show colocation data collected as part of the UK Clean Air program funded QUANT project, and use the tools described above to investigate the impact of the observed errors on end-use.

Figure 5 shows two colocated NO$_2$ measurements, from two different LCS devices using only their out-of-box calibrations (i.e. no colocation data from that site was used to improve performance), compared with colocated reference measurements at an urban background site in the city of Manchester. Unlike the modelled instruments in Sect. 4.1, the combination of error sources is unknown in this case and we can thus only assess the LCS measurement performance through comparison with the reference measurements using metrics and visual tools. There are obvious differences in the performance of both LCS instruments shown in Fig. 5. LCS1 (a-panels) shows an appreciable difference in the time-series baseline, which can be interpreted from both the regression (b_1 <1) and the B-A plots as a proportional bias. This bias also impacts the REU plot, with a minimum in the region where the regression best fit line crosses the 1:1 line (~17ppb). It is worth noting that these plots do not directly identify the source of the proportional bias, with sensor response to the target compound or another covarying compound possible, but provides information on how much it impacts the data. For LCS2 (Fig. 5, b-panels) any proportional bias is significantly smaller, with the B-A plot showing a much more symmetrical distribution of points around the central line across the observed mixing ratio range, although this is not a normal distribution as evidenced by the heteroscedastic nature of the differences, indicating the cause is not entirely random in nature. The lack of a large proportional bias also results in the REU plot showing a continued reduction in relative uncertainty as the true NO_2 concentration increases. Interestingly, both LCS's also show an additional bias at the highest NO_2 values observed. This does not significantly impact the REU, due to its relative nature, but can be seen in the regression and B-A plots. Correcting for the observed proportional bias in LCS1 and LCS2 improves the observed performance by providing the errors with a more symmetrical distribution (LCS1* and LCS2* shown in Fig. S2).

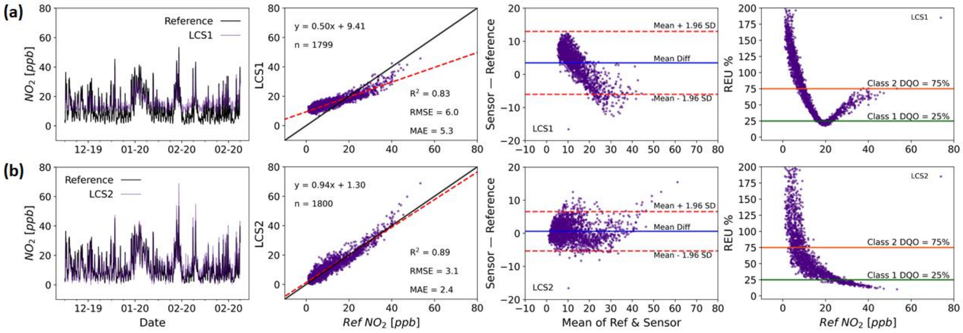

Figure 5. Time series (left panels), regression plots (middle-left panels), Bland-Altman plots (middle-right panels) and REU (right panels; NO_2 Class 1 DQO = 25% & Class 2 DQO = 75%) for NO_2 measurements by two LCS systems of different brands (a and b panels) in the same location (Manchester Supersite, December 2019 to February 2020. Time res 1 h).

Figure 6 shows three out-of-the-box $PM_{2.5}$ measurements made by three devices from the same brand in spring, located at two sites: the first two at an urban background (LCS3 & LCS4, a and b panels) and the third at a roadside (LCS5, c panels). As the regression and the B-A plots show, all LCS measurements in Fig. 6 have a proportional bias compared with the reference, with the LCS over predicting the reference values. Both LCS's at the urban background site show very similar performance, indicating that the devices are similarly affected by errors. This internal consistency is highly desirable, especially when LCS's are to be deployed in networks, as although mean absolute measurement error may be high, differences between identical devices are likely to be interpretable.

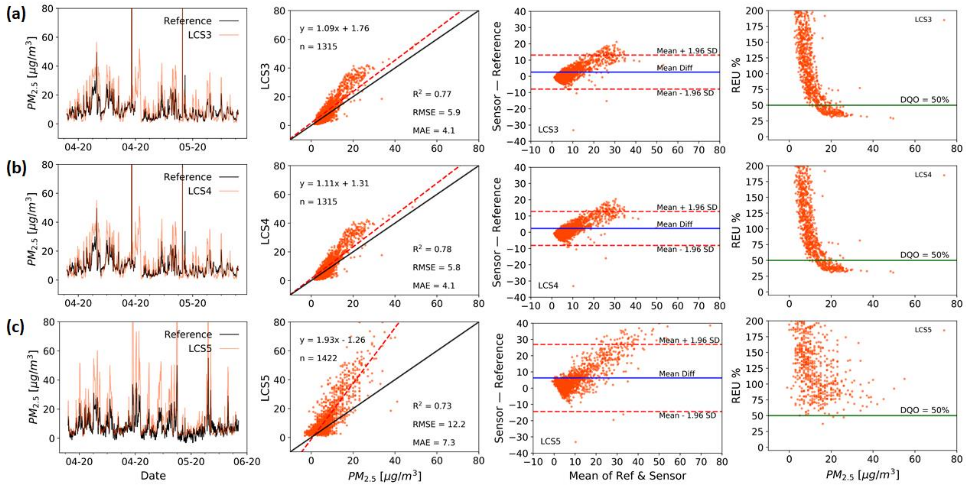

Figure 6. Time series (left panels), regression plots (middle-left panels), Bland-Altman plots (middle-right panels) and REU (right panels, DQO for $PM_{2.5}$ = 50%) for $PM_{2.5}$ measurements by three LCS systems of the same brand (panels a, b and c) in different locations: an urban background (Manchester Supersite, panels a and b) and a roadside site (York, panel c) (April & May 2020, time res 1 h).

The LCS data from the roadside location (LCS5) show significantly lower precision than those at the urban background site, as seen in the B-A plot. This could be caused by differences in particle properties and size distributions between the two sites (Gramsch et al., 2021), and by the high frequency variation of transport emissions close to the roadside side and turbulence effects (Baldauf et al., 2009; Makar et al., 2021). Duplicate measurements show that all sensors of this type responded similarly in this roadside environment (not shown here), supporting the high internal consistency of this device, but indicating a spatial heterogeneity in some key error sources. It is also worth noting that the gold standard instruments at the two sites are not "reference method" but "reference equivalent methods" (GDE, 2010), each using a different measurement technique: while an optical spectrometer (Palas Fidas 200) is used in Manchester, the York instrument uses a Beta attenuation method (Met One BAM 1020), which could also potentially lead to some of the observed differences. The increased apparent random variability for LCS5, combined with the proportional bias, results in significantly higher measurement uncertainty across the observed range, as can be seen by the REU plots, with LCS5 never reaching an acceptable DQO level (50% for $PM_{2.5}$). As with the NO_2 sensors (Fig. 5), if the observed proportional bias is corrected the linearly bias-corrected sensors (Fig. S3) show a much improved comparison with the reference measurement, specially LCS5*. In this case the B-A plot now shows an error characteristic more dominated by random errors, and the REU plots shows a significant reduction, with the REU at 10 ugm^{-3} reducing from ~75 to ~50%.

As a comparison for the LCS data shown above, Fig. 7 shows two identical NO_2 reference grade instruments, Teledyne T200U (Chemiluminescence method) at the Manchester urban background site (panels a and b) at during two different time periods, with a Teledyne T500U (CAPS detection method) used as the "ground truth" instrument. Instrument "a" manifests a significant proportional bias, in contrast to instrument "b", but both show differences that could be non-negligible depending on the application. The deviations observed in instrument "a" was due to the cell pressure being above specification by ~20%, unnoticed while the instrument was in operation.

This demonstrates the importance of checking instrument parameters regularly in the field even if the data appears reasonable.

As the LCS error structure is determined relative to the performance of a reference measurement, if the reference instrument suffers from significant errors this will affect the outcomes of the performance assessment, due to the assumption that all the errors reside with the LCS. As Fig. 7 shows, however, this assumption is not necessarily always valid and potentially argues that reference instruments used in colocation studies should be subject to further error characterisation, including possible colocation with other reference instruments. As a similar comparison of reference instruments, Fig. S4 shows two ozone research grade instruments (a Thermo 49i and a 2B).

It is worth noting that even when using reference, or reference equivalent, grade instrumentation, inherent measurement errors mean that relative uncertainty, as shown in the REU plot, increases asymptotically at lower values. This is not unexpected, but is potentially important as ambient target concentration recommendations continue to fall based on updated health evidence (World Health Organization, 2021).

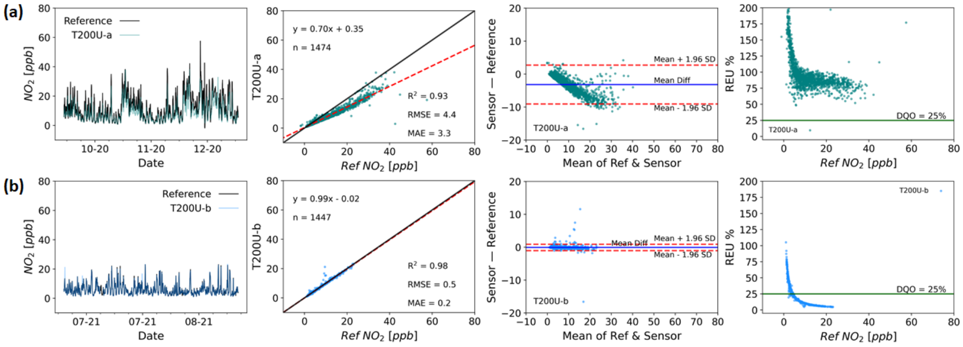

Figure 7. Time series (left panel), regression plots (middle-left panel), Bland-Altman plots (middle-right panel) and REU (right panel, DQO for NO$_2$ = 25%) for two identical (Teledyne T200U) reference NO$_2$ instruments (panels a and b) colocated at the Manchester Supersite (1h time res). The first instrument between October & November 2020 and the second between July & August 2021.

5. Discussion

The widespread use of colocation studies to assess measurement device performance, means many examples exist in the LCS literature where different devices are compared using summary metrics for field or laboratory studies (Broday, 2017; Duvall et al., 2016; Hofman et al., 2022; Karagulian et al., 2019; Mueller et al., 2017; Rai et al., 2017; van Zoest et al., 2019). Although these comparisons do provide useful information, they can be misleading for end users wanting to compare the performance of different devices, as they are often carried out under different conditions and do not present the data or experimental design in full. Even in the case where comparisons have been done under identical conditions, the data still needs to be treated with caution, as inevitable differences between assessment environment and proposed application environment, as well as any changes to instrument/sensor design or data processing, mean that past performance does not guarantee future performance.

All measurement devices suffer from measurement errors, many of which are potentially significant depending on the application, with devices and their error susceptibility covering a broad spectrum. As evidenced by Fig. 7, reference instruments are not immune from this phenomena, with the proportional bias of one of the NOx instruments clearly affecting its measurements resulting in the absolute error increasing with concentration. As the requirements on measurement devices continue to increase, driven in part by new evidence supporting the reduction of air pollutant target values, the devices currently being used for a particular application could no longer be fit-for-purpose in the situation where the limit value has decreased to the point where it is small relative to the device's uncertainty.

Single value performance metrics, such as R^2 and RMSE, can seem convenient when comparing multiple co-located devices as they facilitate decision making when a threshold criterion is defined. However, these scalar values hide important information about the scale and / or distribution of the errors within a dataset; graphical summaries of the measurements themselves can offer significantly more insight into the impact of measurement errors on device performance and ultimate capabilities. Of particular use in air pollution measurements is the ability to see how the errors manifest themselves in relation to our best estimate of the true pollutant concentration, as often applications have specific target pollutant concentration ranges of interest. For example the two NO_2 LCS devices shown in Fig. 5 have similar R^2 values of 0.83 and 0.89, but one is suffering from a strong proportional bias that impacts on measurements either side of the 18ppb crossing-point. Errors, or combinations of errors, frequently result in varying magnitude of the observed measurement inaccuracies across the concentration space observed, and it is often useful to assess both the absolute and relative effects of the errors. By getting a more complete picture of the device performance, decisions can be made on the effectiveness of simple corrections, such as correcting for an apparent proportional bias using an assumption of a linear error model. Ultimately end users need to identify the data requirements a priori and design quantifiable success criteria by which to judge the data. For example, rather than just wanting to measure the 8 hour average NO_2, be more specific and require that this needs to be accurate to within 5 ppb, have demonstrated approximately normally distributed errors in a representative environment for the period of interest, and no statistical evidence of deviation from a linear correlation with the reference measurement over the target concentration range for the period of interest.

A major challenge comes from complex errors, such as interferences from other compounds or with environmental factors, that vary temporally and/or spatially. Similar graphical techniques to those presented above can be used to identify the existence of such relationships, but correcting for them remains a challenge. This brings into question the power of colocation studies, as they can ultimately never be performed under the exact conditions for every intended application. The $PM_{2.5}$ sensors shown in Fig. 6 demonstrate this, as if a colocation dataset generated at the urban background site was used to inform a decision about the applicability of these devices to a roadside monitoring task, then an overly optimistic assessment of the scale of the errors to be expected would be likely. It is therefore always desirable that colocation studies are as relevant as possible to the desired application, and this is even more paramount in the case where the error sources are poorly specified. For this reason, complete meta-data on the range of conditions over which a study was conducted is key information in judging its applicability to different users.

Although there is no strict definition on what makes a device a LCS, we often make the categorization based on the hardware used. Standard reference measurement instruments are generally based on well-characterized techniques developed and improved over years, based primarily on the progressive refinement of hardware (e.g. materials used for the detection elements, electronic circuits to filter noise, refinement of production methods, etc.). Although LCS sensor technologies are improving, it is interesting that many of the significant improvements that have been made to LCS performance have been through software, rather than hardware advances. As more colocation data is generated in different environments, many LCS manufacturers have been able to develop data correction algorithms that minimise the scale of the errors that are present on the LCS hardware. This can greatly improve the performance of LCS devices, and has been a large factor in the improvements seen in these devices over recent years. These algorithms are, however, inevitably imperfect and can suffer from concept drift (De Vito et al., 2020), caused by the lack of available colocation data over a full spectrum of atmospheric complexity. Furthermore, any kind of statistical model introduces a new error source that can work in conjunction with the pre-existing measurement errors to drastically change the observed error characteristics, making it much more difficult for users to interpret and extrapolate from colocation study performance to intended application. If end users are to be able to make well informed decisions about device applicability to a particular task, then an argument can be made for information on the scale of the error corrections made to a reported measurement to be made available, ideally alongside and a demonstration of its benefits in a relevant environment. Unfortunately, this colocation data is costly to generate, meaning relevant data often does not exist, and when it does is often not communicated in such a way that enables the user to make a fully informed decision.

6. Conclusions

In situ measurements of air pollutants are central to our ability to identify and mitigate poor air quality. Measurement applications are wide ranging, from assessing legal compliance to quantifying the impact of an intervention. The range of available measurement tools for key pollutants is also increasingly broad, with instrument price tags spreading several orders of magnitude. In order for a measurement device to be of use for a particular application it must be fit-for-purpose, with cost, useability and data quality all needing to be considered. Understanding measurement uncertainty is key in choosing the correct tool for the job, but in order for this to be assessed the job needs to be fully specified a priori. The specific data requirements of each measurement application need to be understood and a measurement solution chosen that is capable of providing data with sufficient information content.

In order to aid end users in extrapolating from colocation study performance to potential performance in a specific application, performance metrics are often used. Although single value performance metrics do convey some useful information about the agreement between the data from the measurement device being assessed and the reference data, they can often be misleading in their evaluation of performance. This dictates a more rigorous and empirical approach to data uncertainty assessment in order to determine if a measurement is fit for purpose. The ability to assess device performance across the observed concentration range, as in the B-A and REU plots, enables an end-user to make an informed decision about the capabilities of a measurement device in the target concentration range. These visual tools also help identify any simple corrections that can be applied to improve performance. In contrast, if an end-user was only provided with a single value metric, such as R^2 or RMSE then it would be significantly more difficult to understand the likely implications of the measurement uncertainties.

All measurement devices suffer from errors, which result in deviations between the reported and true values. These errors can come from a multitude of sources, with the scale of the deviation from the true value being dependent on the nature of the error. Although a known measurement uncertainty for all applications would be ideal for end users to be able to assess measurement device suitability for purpose, in many cases, especially for LCS, this is not possible due to the presence of poorly characterised, or sometimes unknown, error sources. In the absence of this, useful information on likely measurement performance can be obtained using colocation data compared with a measurement with a quantified uncertainty. It is important that such a colocation study is carried out in an environment as similar as possible to the application environment, as the unknown nature of many error sources means their magnitude can change significantly between different locations and/or seasons (e.g. Fig. 6). Ideally, depending on the measurement task, the user could use the colocation data to model the error causes and use this to develop strategies to minimise final measurement uncertainty. Unfortunately, relevant colocation study data is often not available, and to generate the data would be prohibitively costly, which limits the user's ability to make a realistic assessment of likely uncertainties. The presence of, often complex, error minimisation post processing or calibration algorithms further complicates things. This additional uncertainty is most likely to bias any performance prediction if the end user is unaware of the purpose or scale of the data corrections, and their applicability to the target environmental conditions. Ideally, long term colocation data sets demonstrating the performance of measurement hardware and software, in a range of relevant locations, over multiple seasons, and carried out by impartial bodies would be available to inform measurement solution decisions.

In order for end users to take full advantage of the ever increasing range of air pollution measurement devices available, the questions being asked of the data must be consummate with the information content of the data. Ultimately this information content is determined by the measurement uncertainty. Thus, providing end users with as accurate an estimate as possible of the likely measurement uncertainty, in any specific application, is essential if end users are to be able to make informed decisions. Similarly, end users must specify the data uncertainty requirements for each specific task if the correct tool for the job is to be identified. This requirement for air quality management strategies to acknowledge the capabilities of available devices, both in the setting and monitoring of limits, will only become increasingly important as target levels continue to decrease.

Supplementary

The supplement related to this article is available online.

Code and data availability

The QUANT intercomparison study data will be made publically available once the study has finished, in early 2023. In the meantime, data can be obtained upon request from the corresponding authors.

Author contributions

PE: Funding acquisition; Supervision. SD and PE: Project administration; Formal analysis. SD, PE & SL: Conceptualization; Methodology; Investigation. SD & SL: Visualization; Software. KR, NM, MF: Resources. SD, SL, KR, NM, MF: Data curation. SD, PE, SL, TB, NM, TG & DH: Writing – review & editing.

Competing interests

The authors declare that they have no conflict of interest.

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