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MONTE CARLO MODEL FOR DETERMINING SENSITIVITY OF ANNUALISED MASS EMISSION MONITORING TO KEY UNCERTAINTIES

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Monte Carlo model for determining sensitivity of annualised mass emission monitoring to key uncertainties

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CONTENTS

1 INTRODUCTION

This report details work carried out under the IMPRESS project, Metrology to Underpin Future Regulation of Industrial Emissions. This work is part of the work package on uncertainty of flow and annual emission determination, in particular using Monte-Carlo simulation to assess overall uncertainty in the monitoring of mass emissions from stationary sources.

1.1 STANDARDS AND REGULATION

It is widely accepted that emissions from industrial facilities can represent a danger to the environment and human health. As a result emissions have to be quantified and controlled in order to reduce the risk posed to acceptable levels. In the European Union (EU) emission limits are set out in the Industrial Emissions Directive (IED) (European Parliment, 2010). This legislation sets out a series of emission limit values (ELV) for different pollutant species which relevant industrial sites are required to comply with.

In order to enforce the IED there has to be standards to control how emissions are measured, ensuring that they are measured accurately and consistently across all sites covered by the legislation. The IED sets out the required frequency of emissions monitoring, with some species only requiring periodic measurement while many require constant monitoring with automated measurement systems (AMS).

Each species will have one or more international standards outlining procedures for obtaining suitable measurements. The standard will outline a standard reference method (SRM) (e.g. Ion Chromatography for SO2 as described in EN 14791:2005 (BSI, 2005)) which should be used for monitoring, but alternative methods (AM) can be acceptable for monitoring if they can demonstrate equivalence with the SRM. Demonstrating equivalency of an AM requires comparison with the SRM according to the procedure set out in EN 14793:2005, Stationary source emission – Intralaboratory validation procedure for an alternative method compared to a reference method (BSI, 2005).

In addition to this the AMS must be operated according to a procedure to ensure measurements are consistent and meet uncertainty requirements. Quality assurance procedures for AMS techniques for stationary sources are set out in EN 14181:2014 (BSI, 2014). This consists of four levels of validation testing, quality assurance level 1 (QAL1), QAL2, QAL3 and annual surveillance testing (AST). QAL1 ensures the instrument is suitable for the application and is installed correctly. QAL2 sets out how the AMS should be calibrated with parallel measuring against a reference method. QAL3 checks for and controls drift in the instrument over time using periodic measurements of a reference gas cylinder. AST is a shorter version of the QAL2 to ensure the calibration function is still valid by comparison with a reference method.

These international standards are enforced by local competent authorities, e.g. Environment Agency (EA) in England. Such bodies often produce guidance documents to help operators interpret the standards. An example of this are EA technical guidance notes (e.g. TGN 20 (Environment Agency, 2012)), which outlines how EN 14181 is applied) and method implementation documents (e.g. MID 14181 (Environment Agency, 2014)).

1.2 PREVIOUS STUDIES

Quantifying the uncertainty of these processes is important since the measurements are used to enforce the legislation. However very little attention is paid to the overall uncertainty of the annualised emission totals, with uncertainty assessments tending to focus on one part of the process.

Some people have looked at how EN 14181 is implemented in practice and its ability to produce accurate measurements. Graham (Graham, 2010) warns that the field trials to certify instrument may not be suitable. Product conformity certificates are based on data from 3 month field trials. For the LCP category incinerators are seen as the most demanding environments, so if the field testing was at an incinerator the instrument will be suitable for all types of LCP. Graham points out that incinerators tend to emit low levels of certain substances that may be more common at other emission sites. This could result in an analyser being passed for use when it has not been tested over the full stated range for certain species.

Operators also suggested to Graham that the QAL 2 tests took too long, with tests taking up to a week where multiple components are measured. Given the reliability requirements, a backup system has to be in place to keep within the downtime limits. Graham suggests reducing the number of repeats for each measurement to cut the time taken, but this might negatively influence the quality of the calibration, increasing the required frequency of future calibrations.

Graham also highlights the assumption that the SRM has no bias or uncertainty, something that can cause QAL 2 failures. National guidance in Denmark (FORCE Technology, 2015) requires the investigation of any calibration function where the gradient is outside the 0.8-1.2 range, a sanity check measure that can lead to the identification of a poorly performing SRM or problems with the AMS instrument that might otherwise be missed regardless of whether other functional tests are passed. Graham finds that QAL 3 testing is largely well implemented, but suggests that operators prefer using the Shewhart control charts over the CUSUM charts. Operators would also prefer not to have to implement QAL 3 where instruments have their own internal checks and corrections, as this is seen as repetition with no benefit in terms of reducing error through drift, although this could lead to undetected drift were these systems to fail.

Freibel et al. (Freibel, et al., 2007) investigated how three AMS instruments fared during eighteen months of field trials while operating under EN 14181, with particular attention on the performance of QAL3 testing using the alternative Shewhart and CUSUM control tables. Freibel also compares the principles from the M20 technical guidance note (TGN M20) (Environment Agency, 2012) and the German guidance standard VDI 3950 (VDI, 2006). One area where EN 14181, TGN M20 and VDI 3950 differ is in how they calculate s_{AMS}, the standard deviation used in both types of control tables. EN 14181 states that it should be derived from information obtained for the QAL1 calculations for the zero and span points, while VDI 3950 sets it at 3% of the measuring range. TGN M20 allows s_{AMS} to be calculated from the first ten readings at zero and span during commissioning or the QAL2. The differences in these approaches leads to variation in the sensitivity to drift. Freibel et al. found that the TGN M20 method resulted in s_{AMS} being too small so that measurements are reported as exceeding the limits even when no drift has occurred, so they concluded that using the VDI 3950 guidelines produced the best results.

These assessments help build confidence in the ability of EN 14181 and the other standards to meet the uncertainty requirements of the legislation. However the use of such piecemeal studies could lead to many systematic sources of uncertainty being missed, potentially resulting in underestimation of actual overall uncertainty.

In order to address this issue the Emissions and Atmospheric Metrology group have developed a Monte-Carlo model of an instrument operating under the procedures in EN 14181 over a period of years, representing a full 5-year cycle allowed for AMS at large combustion plants (LCP) under the IED between QAL2 calibrations.

2 MODELLING EN 14181

2.1 MONTE-CARLO SIMULATION AND ITS IMPLEMENTATION IN THE MODEL

MCS involves mathematically modelling a process, then running the model many times with input conditions that are sampled from probably density function (PDF). The PDF for each input parameter is defined according to the individual uncertainties of each input parameter within the model (Equation 1). If there are sufficient repeats within the model run then the spread of the output will represent the range of potential results that can be achieved with those input parameters and their associated uncertainties. The standard deviation of the results of a model run is therefore an indication of the overall uncertainty of the whole process.

$$R = M + e1 + e2 + \ldots \qquad \text{Equation 1}$$

Where:

R	Realised value in the model
M	Measurand value
e1, e2, …	Error values from different sources, sampled from PDF for each source

The input parameters are sampled from a PDF representing the measurement error on the input. In general this is a Gaussian (normal) distribution (Equation 2), although detection limit error is an exception to this and is sampled from a uniform distribution with the instruments reported detection limit as the limit values on the distribution.

$$f(x \mid \mu, \sigma^2) = \frac{1}{\sqrt{2\sigma^2\pi}}\, e^{-\frac{(x-\mu)^2}{2\sigma^2}} \qquad \text{Equation 2}$$

Where:

μ	Mean of the distribution, generally zero unless the error is systematic
σ	Standard deviation of the probability density function, i.e. the uncertainty for the error

Assessment and combination of the uncertainties follows the rules in the BIPM Guide to Uncertainty in Measurement (GUM) (JCGM, 2008). When talking about uncertainties GUM defines measurand as the actual value being measured and realised value as the result of the measurement that has been influenced by the uncertainty. More details on using MCS to investigate propagation of uncertainty can be found in GUM supplement 1 (JCGM, 2008).

2.2 MODEL IMPLEMENTATION

The model represents all the processes involved in running an AMS operating under EN 14181, i.e. measurements and quality assurance regime. The model simulates the processes involved in the measurement and quality assurance regime of an instrument over a period of up to five years (Figure 1), indicating the level of error expected in that period. Each measurement made with the instrument will have associated uncertainties, e.g. error from the offset between measurement and calibration temperatures.

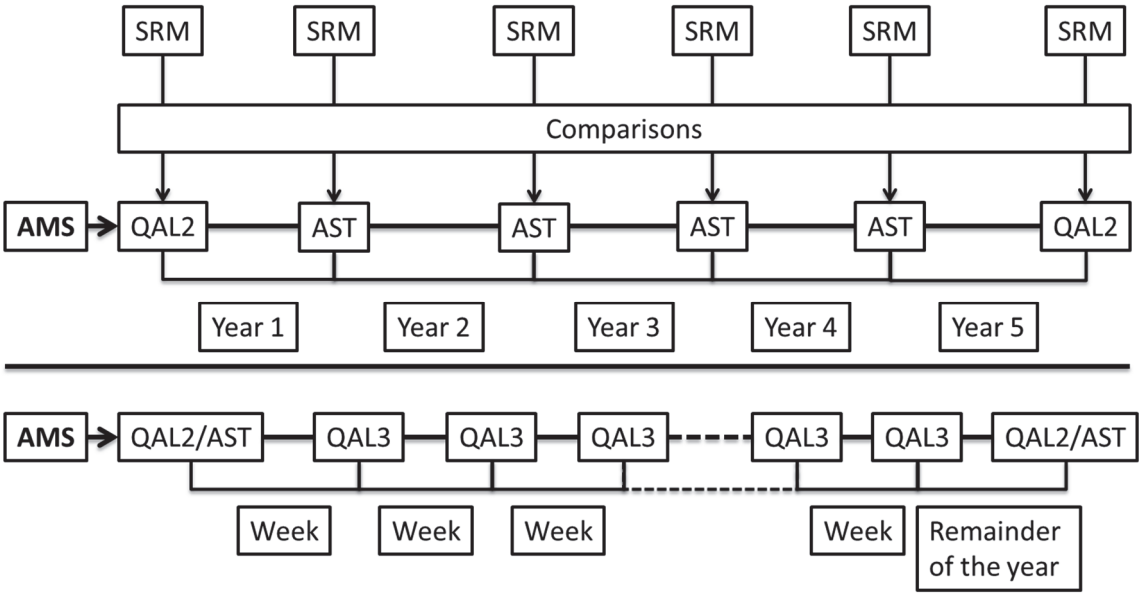

Figure 1 Frequency of quality assurance testing. Top section shows the required frequency of tests involving intercomparison with an SRM. Lower section indicates an example of QAL3 frequency, but the timing is based on the product conformity certification so the week shown here is just an illustrative example

There are two classifications of sources of uncertainty: systematic and random. Random sources of uncertainty will change with every repeat of the measurement, but if the measurement was repeated enough times the resulting error would average out. In contrast systematic sources of uncertainty will affect measurements in the same way over time, an offset from the measurand that will not average out over multiple measurements of the same thing.

Within the model an example of a systematic error would be if the AMS is not calibrated perfectly, so all measured values made with it will be offset from the measurand by the same amount. The repeatability performance characteristic is an example of a random error that would average out when taken over a large number of repeated measurements. The scale of each variation in the model is set by the uncertainty relating to the particular action as described in the product conformity certificate for the instrument. Each measurement made by the model instrument will therefore include random terms for uncertainty relating to cross sensitivity, temperature dependent shift, zero and span drift, etc., so that all sources of error (systematic and random) are included. The measurand value is computed alongside the repeated realised values so that the MCS results can be compared, giving an indication of the scale of errors propagating during operation of the instrument while it is operated under EN 14181.

The sources of uncertainty within the process are encapsulated within the model, so for each repeat in the MCS the measurement input parameters are adjusted by a random error, the magnitude of which is controlled by the uncertainty of the operation. This process produces a range of possible outcomes, which with enough repetitions will incorporate the majority of potential results. The distribution of the outcomes indicates the uncertainty in the model output.

The model has been created using the R programming language (R Core Team, 2014) which is well suited to statistical requirements of MCS.

2.3 SCOPE OF THE MODEL

In a simple case the measured concentration might be affected by just the measurement temperature. This would lead to the measured concentration being a function of the actual concentration and the temperature (Equation 3), with the temperature effect being a function of the difference in temperature

at the time of the calibration and the measurement (Equation 5) and the sensitivity to that temperature difference (Equation 4).

$$R = f(c) + f(T) \quad \text{Equation 3}$$

$$f(T) = \Delta T * s \quad \text{Equation 4}$$

$$\Delta T = T_{cal} - T_{meas} \quad \text{Equation 5}$$

Where:

R	Realised value of the concentration
f(c)	Function of concentration
f(T)	Function of temperature
ΔT	Temperature difference between the measurement and calibration
s	Sensitivity, i.e. how temperature will affect the result
T_{meas}	Temperature when the concentration measurement is made
T_{cal}	Temperature when the instrument was calibrated

This forms a simple MCS representing one instrument measuring the concentration. The scope of the model can be changed by altering which variables have PDF applied to them. In the example above, if a PDF was applied to the sensitivity, so for every measurement it was different, then the scope of the MCS would go from a single instrument to a population of instruments measuring the same concentration. The model contains many different PDF, so by choosing which to apply the model can easily investigate a wide variety of different situations.

2.4 MODEL ASSUMPTIONS

The model is based on the real use of a gas analyser instrument under EN 14181, but, like any model, it has to include some assumptions in order to function. Some aspects of the testing under EN 14181 are assumed and not specifically modelled (e.g. the linearity test that is part of the AST). This can be justified since they relate to constant sources of error which will not drift over time, so the offset will be included within a standard calibration. The model takes 30 minute average measurements, assuming that there is no downtime, including timeless quality assurance testing, so every measurement is made regardless of procedural requirements (e.g. running AST). This is obviously a major abstraction compared to real field measuring where downtime can represent ten percent or more of a data set, but is a justified assumption as it has no effect on the uncertainty of the AMS results that are covered within the measurement period.

Uncertainty from cross interference with other substances in the measured flow is included in the model, but with some limitations. Up to three potential sources of cross-interference for the AMS can be included, with the measurand values of these substances provided to the model as inputs. Errors for each interference source are calculated separately and added together to form the total cross interference error on an individual measurement. The errors are randomised based on data provided for the intensity of interference from each interferent, as stated in the instrument test report produced from instrument certification under EN 15267-3 (BSI, 2007). No uncertainties are applied to the concentration of the interfering substance in order to avoid over-complicating the model. Additionally the SRM measurements for parallel running just use an error term, rather than an error scaled by the magnitude of interfering materials. The SRM technique is likely to be influenced by different species, so to limit model complexity this simplification has been made.

The model includes the effect of ambient measurement temperature compared to the ambient temperature at calibration. Example data from measured temperatures could be used if available, but during development and testing of the model assumed temperature data was used. Measurement temperatures are based on a simple sine curve to give some seasonal variation. Random variation is then applied to this seasonal curve for individual measurements with a filter to prevent very high jumps in temperature. Temperature was the only aspect of the weather considered to be within the scope of the model, so any effects of wind or precipitation there might be are not incorporated. This is

justified as most instruments are deployed in an enclosure so will not be affected by external conditions like wind and rain.

In order to reduce the required runtime and simplify the programme the QAL2 does not include the checking for and removal of outliers. The model can be provided with higher N values than the minimum fifteen to represent additional parallel measurements carried out in case of outliers, but correctly the model will not look for them or exclude them. All parallel measurements in the model are used in selection of the calibration function method (a, b or c) and subsequent calculations.
The model currently does not cover all potential methodologies that are allowed under EN 14181. For monitoring drift and precision of the instrument EN 14181 outlines the use of Shewhart, exponential weighted moving average (EWMA) and CUSUM control charts. The Shewhart and CUSUM approaches were looked at by Freibel et al. who found they could give false drift signals when close to the set point. Since both were affected one methodology was chosen to be implemented in the model, the CUSUM control charts. This allows adjustments to be applied to reduce the need for additional calibrations.

The standard deviation of the AMS (s_{AMS}), which is used in QAL 3 testing, is derived from the uncertainties taken from the product conformity certificate rather than being "calculated considering plant conditions". The data in the product conformity certificate is partly derived from field measurements so the stated values should in many cases be suitable assuming the testing was at a similar facility (e.g. LCP). This will not always be the case since LCP and WIP have different emissions profiles, so the use of the default instrument product conformity certificate values will introduce additional uncertainty that the model will not account for. The UK TGN M20 allows the operator to derive s_{AMS} at the start of a QAL2 using measurements of zero and span gases, while also mentioning a simpler approach proposed by the French standards body AFNOR. The AFNOR approach (AFNOR, 2013) calculates s_{AMS} as a fraction of the allowable uncertainty for the species. The uncertainty from s_{AMS} is difficult to quantify, but it is expected to be small and unlikely to significantly impact the overall results of the model, so the use of this assumption is valid. The current s_{AMS} implementation from product type testing could be replaced with alternative met method in future to test the suitability of each solution.

Under normal operation LCP have a calibration range under which their QAL2 is valid and the operator has to monitor exceedances of this limit. The model does not include this monitoring and assumes the calibration algorithm will be valid for all measurements recorded.

The input parameters within the model are summarised in Annex 1.

2.5 TESTING AND VALIDATION

Extensive testing of the model was required to ensure that it both functioned as intended and that the implementation was able to reproduce real results. Model testing was split into several stages, using dummy data to test functionality, then real data to demonstrate model validity.

2.5.1 Dummy data testing

During development each section of the model was tested with simplified dummy data (e.g. constant emission rates, etc.). This allowed the output of each section to be checked against the calculated value for the data to ensure accuracy. This was done for each section independently and repeated once each section had been integrated into the overall model. The full model was run with various constant emission levels which were compared with calculated emission values.

The dummy data tests were all completed successfully with the calculated values matching the model output. Figure 2 shows the results of a run with the AMS measuring a constant 25mg/Nm^3 emission over a five year period with quarterly QAL3 testing. The time series plots show the mean results of the

MCS, while the red lines and shaded area represents +/- 2 standard deviations, i.e. the 95% confidence interval.

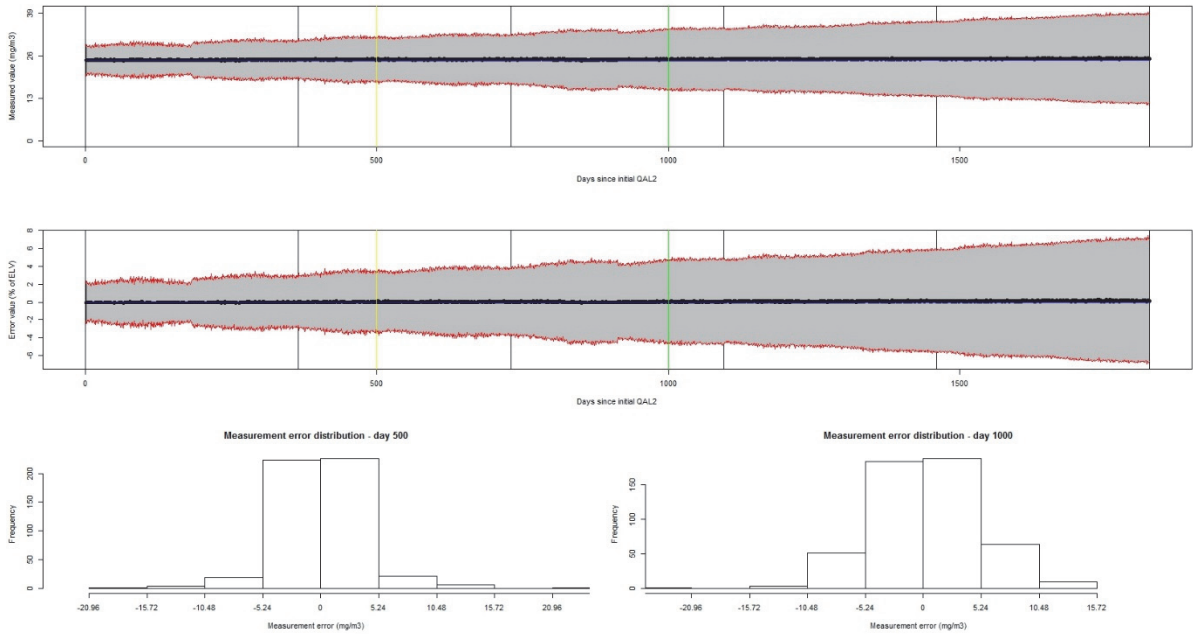

Figure 2 Concentration values with k=2 uncertainty indicated by the shaded area. Top plot is the measured value, while the middle shows the error value as a percentage of ELV. Lower plots give a snapshot of the distribution of errors after 500 and 1000 days in the simulation

Since emissions are often reported as mass emissions, the model has a flow module to represent a flow meter operating under the relevant standard (EN 16911) (BSI, 2013). This allows the model to provide readings of the annual mass emission together with an uncertainty, which matched the expected values for the test dataset.

2.5.2 Real data testing

Once all aspects of the model had been integrated and successfully tested with the dummy data the model was run with real world data. This included full tests to check the model was able to replicate the expected emission totals from real data, but also standalone testing of important modules in the model. In particular the focus was on the QAL2 testing as this sets the calibration function.

Real data from QAL2 tests at two stacks was used to verify that the model was able to reproduce the same results. By setting all the variability in the model to zero every repeat in the MCS will be the same and should match the result from the actual QAL2 that was performed.

Standards for measuring each species (e.g. EN 14791:2005 (BSI, 2005) for SO2) set out minimum uncertainty requirements for SRM uncertainty (e.g. 20% for the SO2 SRM). The suitability of this requirement was tested by using the QAL2 module. The same test was performed with the data, but varying SRM uncertainty to demonstrate how that would alter the likelihood of failing the QAL2 test. As uncertainty on the SRM measurements increases so too does the chance that the AMS and SRM measurements will diverge far enough to fail a QAL2. The model counts the number of attempts required to pass the QAL2 in each repeat. In general this should be one unless the error is sufficient to generate failures. The model was run with 1000 repeats at varying SRM uncertainty with all other variability in the model set to zero.

The main output from the QAL2 testing is the calibration function. The model was set up with no variation in the input parameters and run with data from two QAL2 tests carried out at real sites. The calibration from the model should match the one produced from the field measurements. The

calibration factor from the model is plotted, along with the measurements, as described in EN14181. This result was consistent across all repeats as would be expected with no error applied and also matched the calibration function from the QAL2 the data was from, thus verifying the ability of the model to accurately reproduce real world results.

The tests with real data and modified levels of SRM uncertainty used a different output of the model, counting the number of QAL2 tests required to pass. The real data being used was from a successful QAL2 test where the variability test had been passed and the calibration function adopted for use, so the modelled test would be expected to pass first time in all repeats. As uncertainty is applied to the measurements there is a chance they will become bad enough that it will fail and have to be retested, which the model implements by "re-measuring", generating a new set of realised values. The results of 1000 repeat runs at different SRM uncertainties (Figure 3) show that SRM uncertainty has to rise over 15% before it is sufficient to trigger a significant number of failures.

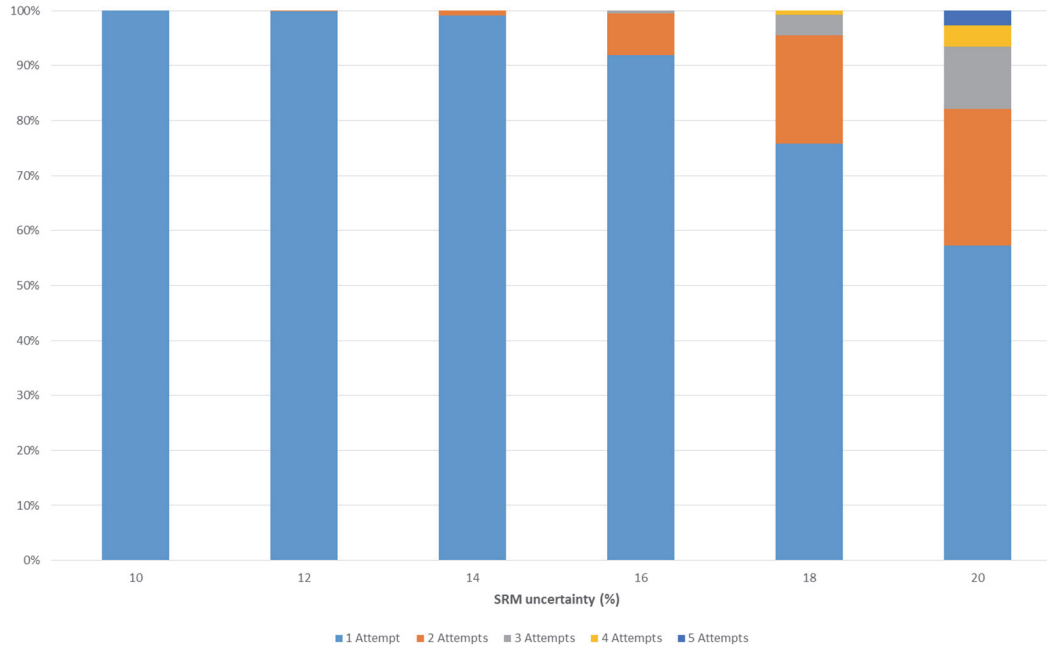

Figure 3 Proportion of repeats where more than one attempt is required to successfully pass a QAL2. Blue indicates passing first time while other colours indicate increasing number of tests required to pass. Rate of failures increases above 15% SRM uncertainty

2.5.3 Conclusions from testing and validation

The dummy and real data tests both showed that the model is functioning as expected and is able to match real world results, including generation of calibration functions.

For measuring SO2 the standard is EN 14791:2005 (BSI, 2005) and this includes the requirements for SRM uncertainty, which can be up to +/-20% at the ELV. From the results presented in Figure 3 it is clear that uncertainty in the SRM can alter measurements enough to cause significant levels of QAL2 failure at uncertainties below 20% uncertainty. In addition to this, for the results presented here, all other potential sources of variation are excluded. When other sources of uncertainty are included in the calculation failure rates are likely to increase. Given that the SRM is assumed to be an unbiased indicator of the true value, the allowable uncertainty should not be sufficient to trigger these failures. These results suggest that the limit on allowable SRM uncertainty needs to be reduced.

Running the QAL2 module with real data, but altering the SRM uncertainty is a first look at what new understanding of uncertainty in the process can be achieved using the model.

3 SENSITIVITY TESTING EN 14181

3.1 METHODOLOGY

In order to assess the relative importance of each uncertainty attribute it is necessary to test them individually. To achieve this the model is run repeatedly with the same data and all attributes set to zero, other than the one under test. That way the resulting overall uncertainty is all from the tested attribute and how its uncertainty manifests itself through the process. The model will be run with the test attribute at a representative range of values (Table 1) to assess the overall impact. The sensitivity of the attribute will depend on the comparative scale of the impact on the overall uncertainty. All tests are assumed to be measuring a constant stream of the pollutant at levels close to the ELV, while measurement temperatures have seasonal variation compared to an initial calibration temperature of 15°C.

Table 1 Tested attributes with the range over which they are tested

Attribute/Influence factor	Test range
SRM uncertainty	0 – 25%
AMS repeatability	0 – 10%
AMS linearity	0 – 10%
AMS cross sensitivity (single species)	0 – 10%
AMS cross sensitivity (three species - complimenting)	0 – 10%
AMS cross sensitivity (three species - conflicting)	0 – 10%
AMS detection limit	0 – 10%
AMS temperature offset from calibration – zero	0 – 10%
AMS temperature offset from calibration – span	0 – 10%
AMS zero drift	0 – 10%
AMS span drift	0 – 10%

For cross sensitivity the model allows up to three different interfering species to be included. The species are defined with two attributes providing the impact on the result and the amount of the substance that needs to be present to illicit that response. All tests will be run with the quantity of the species set to the full response value, with the test range representing the magnitude of that response. The test will be repeated with one and all three species set to provide a response.

The response to the difference in temperature between calibration and the measurement has different magnitudes between zero and span. The sensitivity of these two attributes will therefore be tested separately. Similarly over time the zero and span points will drift from their calibrated level at different rates so they will also be tested separately.

The model is set up as an instrument measuring SO_2 concentration on a stack with a cross sectional area of $2m^2$. Flow rate is constant at 25m/s and the flow rate sensor has monthly QAL3 testing compared to the quarterly QAL3 for the concentration instrument. AST and QAL2 testing are assumed to occur at the same time for both flow and concentration instruments.

3.2 RESULTS

Results are presented for each tested attribute. Overall uncertainties have been recorded for the concentration and mass emission totals. These change over the five years of each run for some variables, so along with the overall value for each test a time-series of measurement standard deviations shows this evolution.

3.2.1 SRM uncertainty

The range of uncertainties is based on the requirement in the reference standard for SO_2 (EN 14791:2005 (BSI, 2005)) that the SRM has to meet a limit on overall uncertainty of ±20% relative at the ELV. Investigating both when the SRM is meeting and exceeding this limit will provide an indication of the suitability of the current limit in EN 14791 and if it should be raised or lowered.

Table 2 Model results indicating overall uncertainties when varying SRM uncertainty

SRM Uncertainty (%)	Concentration overall uncertainty (mg/30 minutes)	Highest recorded concentration uncertainty (mg/30 minutes)	Mass emission overall uncertainty (t/year)
0	0	0	0
1	0.0655	0.0655	0.103
2	0.1305	0.1305	0.206
3	0.2008	0.2008	0.317
4	0.2546	0.2546	0.401
5	0.3226	0.3226	0.509
6	0.4128	0.4128	0.651
7	0.4779	0.4779	0.754
8	0.5214	0.5214	0.822
9	0.6141	0.6141	0.968
10	0.6591	0.6591	1.039
11	0.7245	0.7245	1.142
12	0.8056	0.8092	1.27
13	0.8781	0.8798	1.385
14	0.9008	0.9038	1.42
15	0.9978	1.0045	1.573
16	1.0819	1.1082	1.706
17	1.1625	1.183	1.833
18	1.2007	1.2318	1.893
19	1.2774	1.2909	2.014
20	1.3181	1.3353	2.078
21	1.3864	1.4089	2.186
22	1.4725	1.5009	2.322
23	1.5288	1.5549	2.411
24	1.5984	1.6202	2.52
25	1.6807	1.7354	2.65

The relationship between SRM uncertainty and overall uncertainty is strongly linear (R^2=0.9994), exhibiting very few points of divergence both under and above the allowable limit value set out in EN 14791 (Figure 4).

Since the SRM is only involved in setting the calibration function it will only change when a QAL3 or AST test is failed and the calibration function has to be re-evaluated. ASTs occurring at the same time in each MCS repeat lead to clusters of failures at the end of a year, resulting in step changes in the measurement uncertainty (Figure 5).

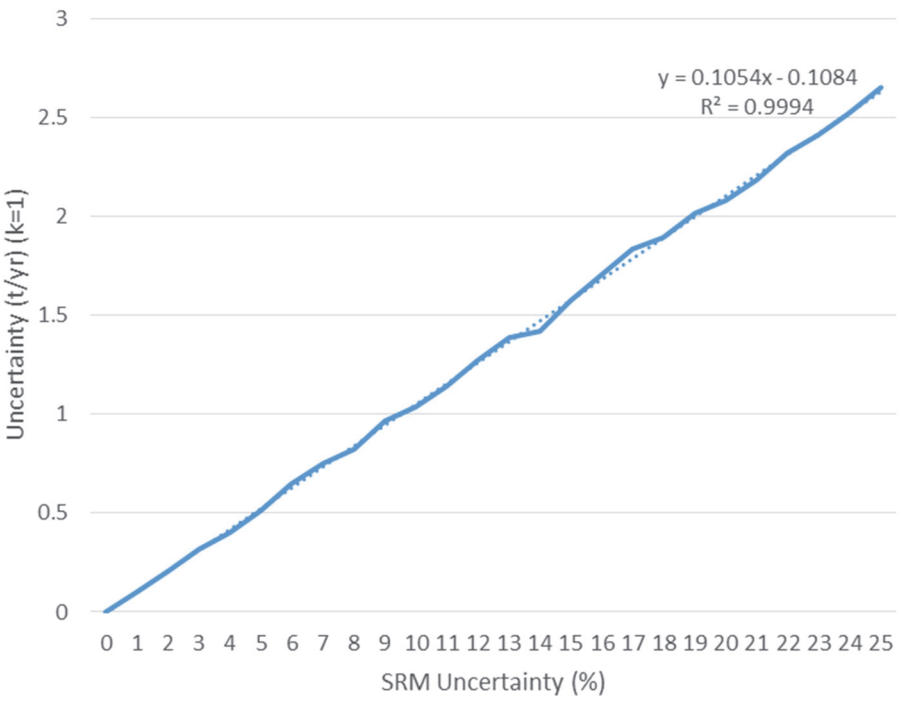

Figure 4 Overall uncertainty of annualised mass emissions when SRM uncertainty is increased

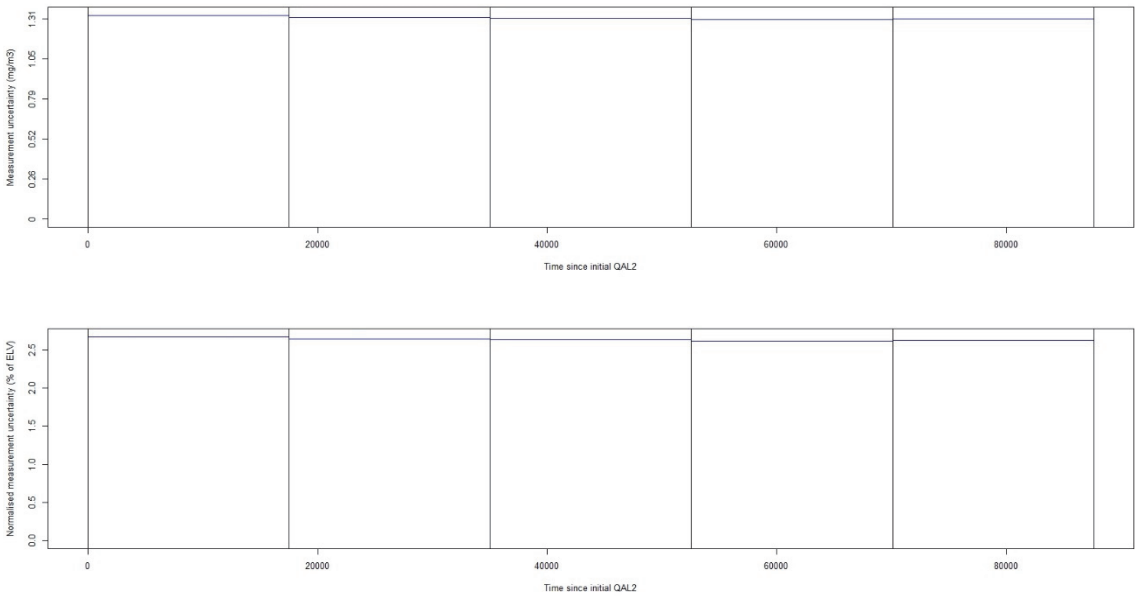

Figure 5 Concentration measurement uncertainty for each measurement after the initial QAL2 in mg/m3 (top) and as a percentage of the ELV (bottom). Vertical lines indicate end of year AST testing. Plot is from a run with SRM uncertainty set to 25%

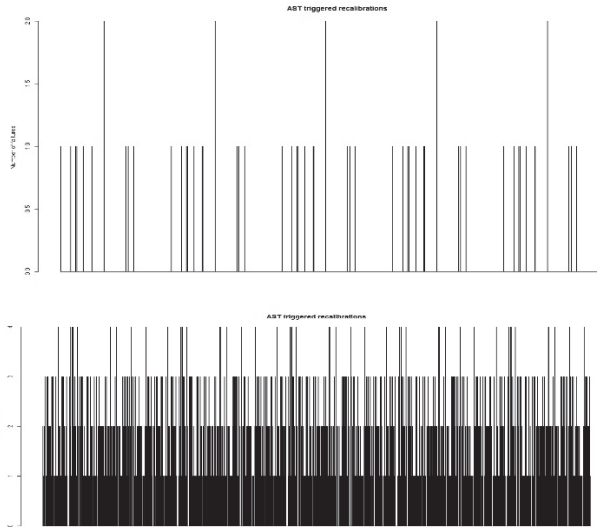

Figure 6 AST failure rate over a five year period at SRM uncertainty of 15% (top) and 25% (bottom). Each bar is one of the 1000 repeats in a model run

3.2.2 AMS repeatability

Repeatability is a random error so the uncertainty varies between each measurement (figure 7), in contrast with the SRM results (Figure 5) which affect the calibration creating a systematic effect. The highest overall uncertainty recorded for a measurement was correlated linearly with increasing repeatability uncertainty (Figure 8), but the overall uncertainty for the overall emissions was more suited to a third order polynomial function (Figure 9).

Table 3 Model results indicating overall uncertainties when varying the AMS repeatability uncertainty characteristic

AMS repeatability (%)	Concentration overall uncertainty (mg/30 minutes)	Highest recorded concentration uncertainty (mg/30 minutes)	Mass emission overall uncertainty (t/year)
0.23	0.1236	0.1310	0.1746
1	0.5387	0.5708	0.7634
2	1.0731	1.1426	1.5338
3	1.6244	1.7316	2.2975
4	1.9768	2.2690	2.7298
5	2.1543	2.7988	2.7970
6	2.3136	3.4834	2.8511
7	2.4782	3.9498	2.8714
8	2.7465	4.4193	3.1260
9	3.0157	5.0273	3.3807
10	3.3675	5.6455	3.6867

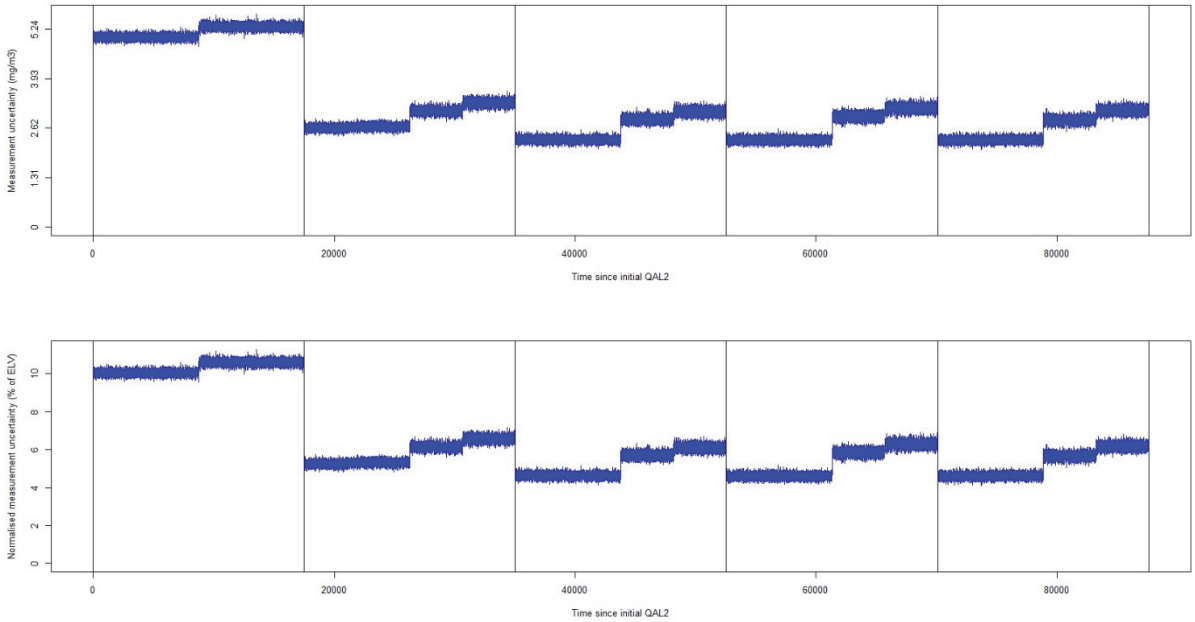

Figure 7 Concentration measurement uncertainty for each measurement after the initial QAL2 in mg/m^3 (top) and as a percentage of the ELV (bottom). Vertical lines indicate end of year AST testing. Plot is from a run with AMS repeatability set to 10%

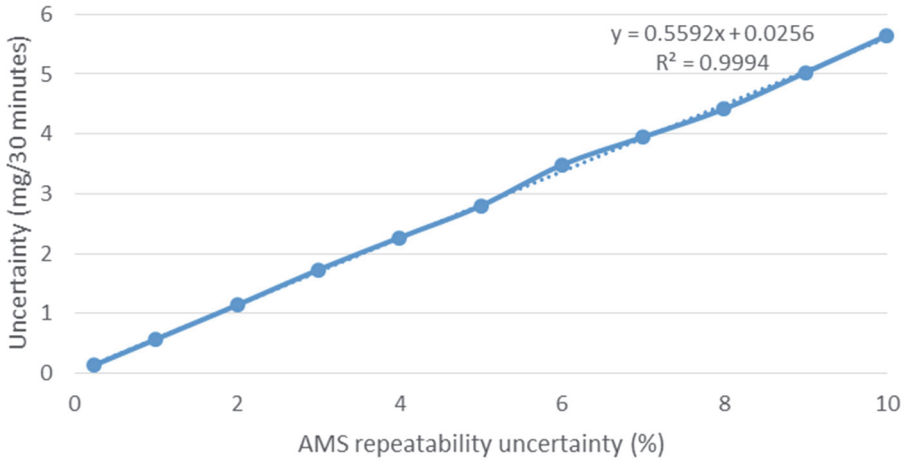

Figure 8 Maximum uncertainty for a single measurement at varying levels of AMS repeatability. Relationship with overall uncertainty is strongly linear over the whole tested range

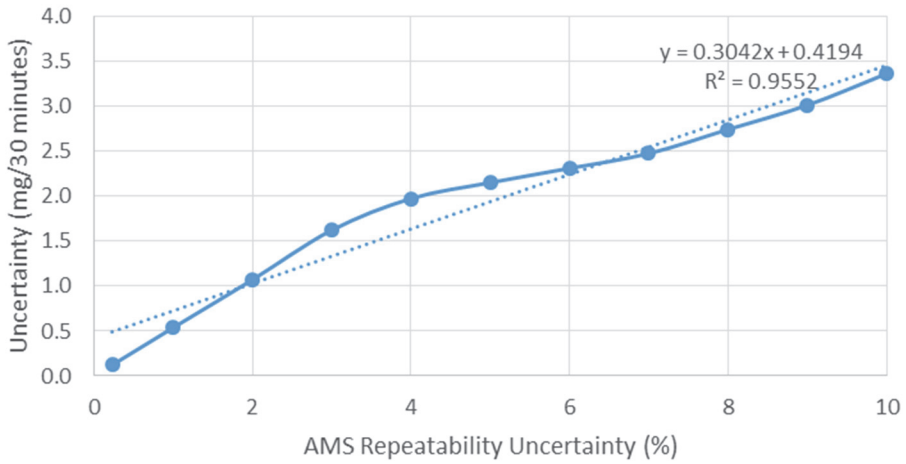

Figure 9 Overall uncertainty in concentration measurements at varying levels of AMS repeatability uncertainty. Less strongly linear but still has R^2>0.95

3.2.3 AMS linearity

Linearity error is due to measurements not precisely mapping to the calibration function. The error is random so varies from one measurement to the next (Figure 10). The results are summarised in Table 4.

Table 4 Model results indicating overall uncertainties when varying the AMS linearity uncertainty characteristic

AMS linearity (%)	Concentration overall uncertainty (mg/30 minutes)	Highest recorded concentration uncertainty (mg/30 minutes)	Mass emission overall uncertainty (t/year)
0.9	0.2163	0.2495	0.08433
1	0.2400	0.2738	0.09244
2	0.5139	0.6136	0.32184
3	0.8255	0.9812	0.67113
4	1.1486	1.4052	1.05295
5	1.4550	1.7229	1.34966
6	1.7507	2.0615	1.62097
7	2.0235	2.4578	1.81026
8	2.3165	2.8317	2.00329
9	2.5477	3.1493	2.10087
10	2.7391	3.3735	2.19876

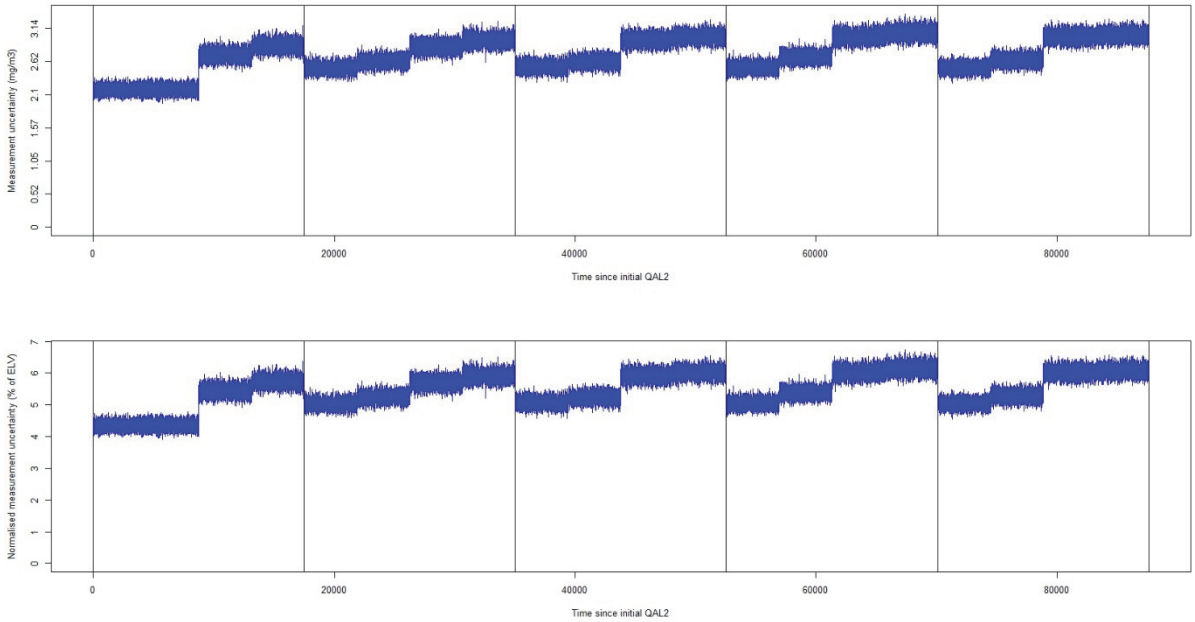

Figure 10 Concentration measurement uncertainty for each measurement after the initial QAL2 in mg/m^3 (top) and as a percentage of the ELV (bottom). Vertical lines indicate end of year AST testing. Plot is from a run with AMS linearity uncertainty set to 10%

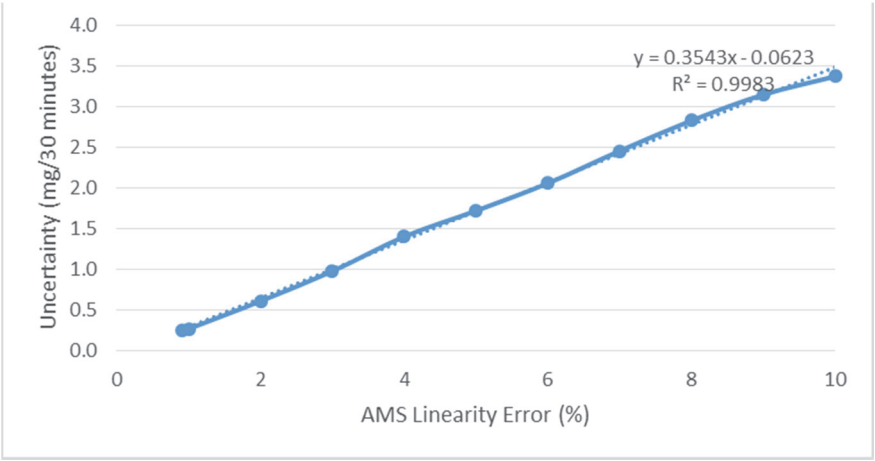

Figure 11 Maximum uncertainty for a single measurement at varying levels of AMS linearity. Relationship with overall uncertainty is strongly linear (R^2=0.9983) over the whole tested range

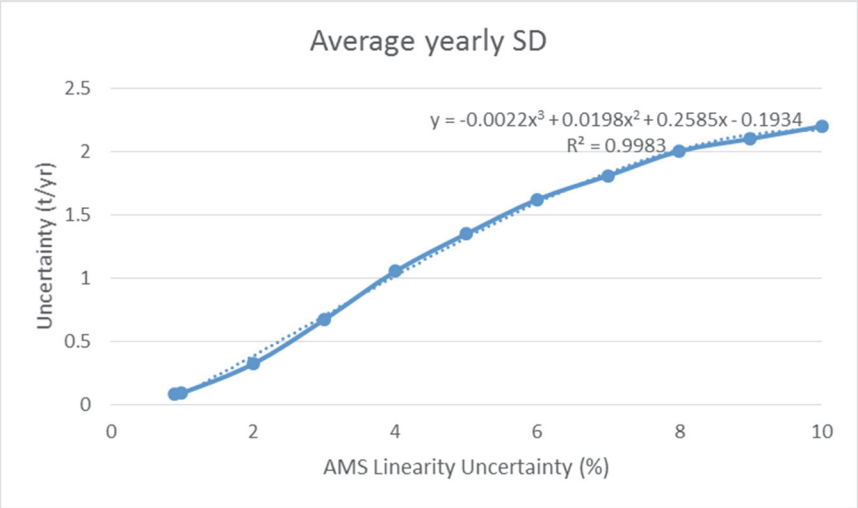

Figure 12 Overall uncertainty in annual mass emissions at varying levels of AMS linearity uncertainty. Curve fitted is a 3rd order polynomial as linear regression produced a sub-optimal fit

3.2.4 AMS cross sensitivity

Three sets of results for cross sensitivity, the first with a single species and two with a spectrum of three cross interfering species all increasing their impact at the same rate. The first three species results have the three sharing the same tendency, so the maximum magnitude will be demonstrated, while the second will feature conflicting tendencies which should cancel out some of the effects. All tendencies are increased at the same rate for all species and the concentrations of the species are kept constant for all tests.

Table 5 Model results indicating overall uncertainties when varying the response to a single interfering species

AMS cross-sensitivity: 1 species (%)	Concentration overall uncertainty (mg/30 minutes)	Highest recorded concentration uncertainty (mg/30 minutes)	Mass emission overall uncertainty (t/year)
1	0.2390	0.2707	0.09716
2	0.4864	0.5490	0.19725
3	0.7177	0.8061	0.29042
4	0.9815	1.1223	0.38232
5	1.1477	1.2939	0.44446
6	1.3913	1.5792	0.55434
7	1.6205	1.8352	0.64183
8	1.7891	2.0426	0.72679
9	1.9798	2.2472	0.75616
10	2.2366	2.5337	0.88137

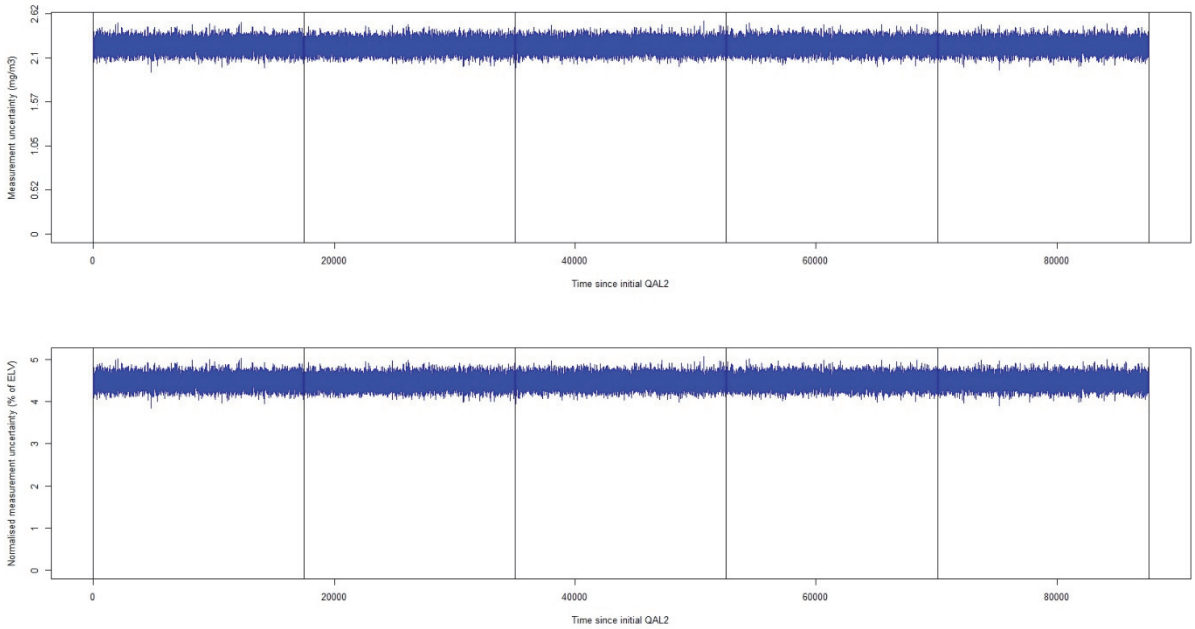

Figure 13 Concentration measurement uncertainty for each measurement after the initial QAL2 in mg/m^3 (top) and as a percentage of the ELV (bottom). Vertical lines indicate end of year AST testing. Plot is from a run with response to a single interfering species set to 10%

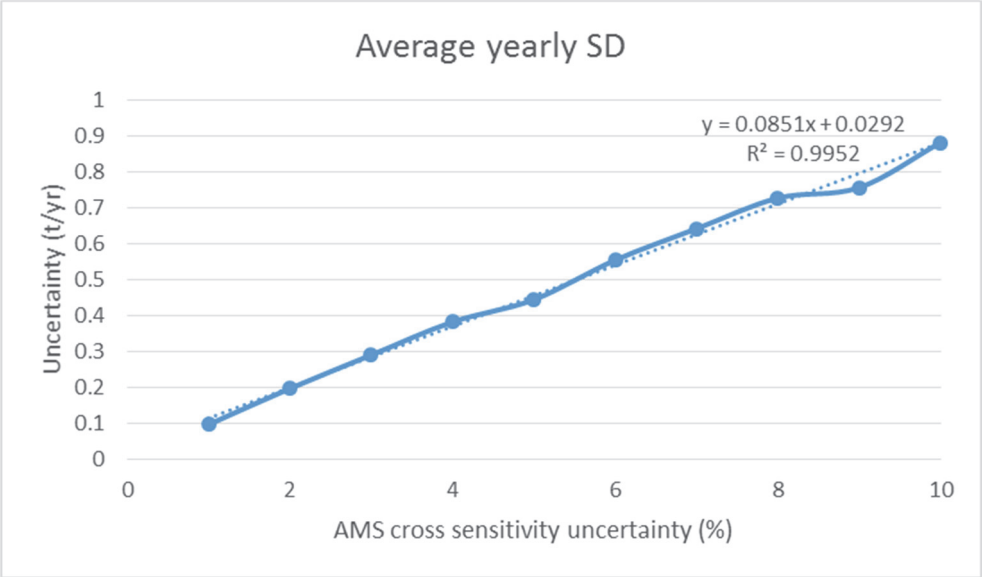

Figure 14 Overall uncertainty in annual mass emissions at varying levels of AMS cross sensitivity to one species

Table 6 Model results indicating overall uncertainties when varying the response to three interfering species with the same tendency

AMS cross-sensitivity: 3 species (%)	Concentration overall uncertainty (mg/30 minutes)	Highest recorded concentration uncertainty (mg/30 minutes)	Mass emission overall uncertainty (t/year)
Typical values (1/1.35/1.4)	0.5328	0.5903	0.2060
1 (1/1/1)	0.4232	0.4721	0.1674

2 (2/2/2)	0.8222	0.9155	0.3316
3 (3/3/3)	1.2269	1.3627	0.4819
4 (4/4/4)	1.6232	1.7927	0.6259
5 (5/5/5)	2.0105	2.2409	0.8151
6 (6/6/6)	2.3536	2.6322	0.9609
7 (7/7/7)	2.6767	2.9980	1.0653
8 (8/8/8)	2.9419	3.2578	1.1801
9 (9/9/9)	3.2168	3.5852	1.3234
10 (10/10/10)	3.4597	3.8029	1.3636

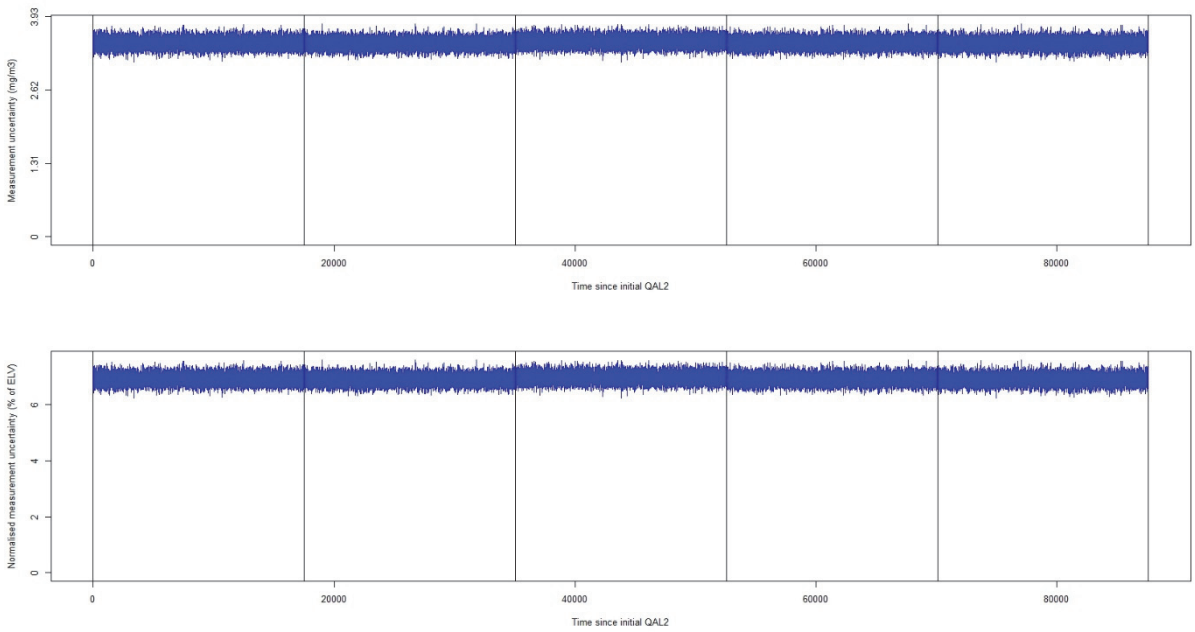

Figure 15 Concentration measurement uncertainty for each measurement after the initial QAL2 in mg/m^3 (top) and as a percentage of the ELV (bottom). Vertical lines indicate end of year AST testing. Plot is from a run with response to three interfering species with the same tendency set to 10%

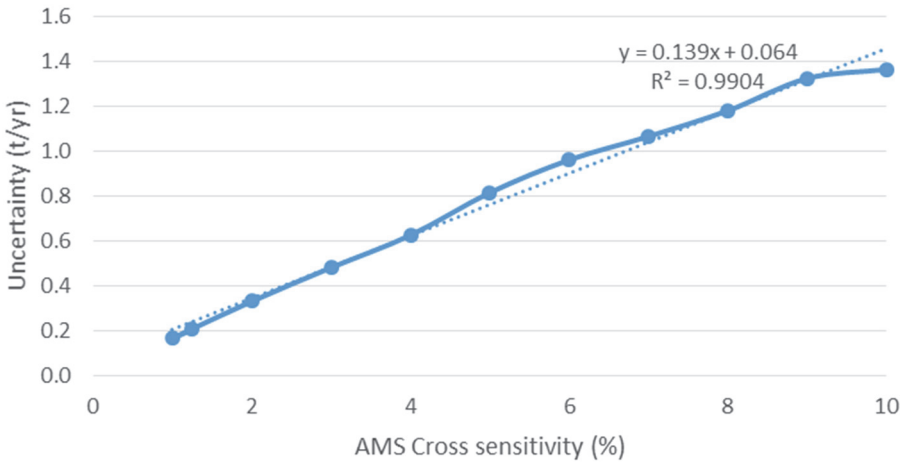

Figure 16 Overall uncertainty in annual mass emissions at varying levels of AMS cross sensitivity to three species with similar types of effect

Table 7 Model results indicating overall uncertainties when varying the response to three interfering species with opposing tendencies

AMS cross-sensitivity: 3 conflicting species (%)	Concentration overall uncertainty (mg/30 minutes)	Highest recorded concentration uncertainty (mg/30 minutes)	Mass emission overall uncertainty (t/year)
Typical values (1/-1.35/-1.4)	0.5317	0.5947	0.2139
1 (1/-1/-1)	0.4211	0.4704	0.1685
2 (2/-2/-2)	0.8428	0.9477	0.3393
3 (3/-3/-3)	1.2244	1.3717	0.4916
4 (4/-4/-4)	1.6034	1.7920	0.6292
5 (5/-5/-5)	2.0059	2.2210	0.8054
6 (6/-6/-6)	2.3473	2.5683	0.9034
7 (7/-7/-7)	2.6623	2.9572	1.0526
8 (8/-8/-8)	2.9476	3.2984	1.1716
9 (9/-9/-9)	3.2107	3.5719	1.2652
10 (10/-10/-10)	3.4724	3.8463	1.3961

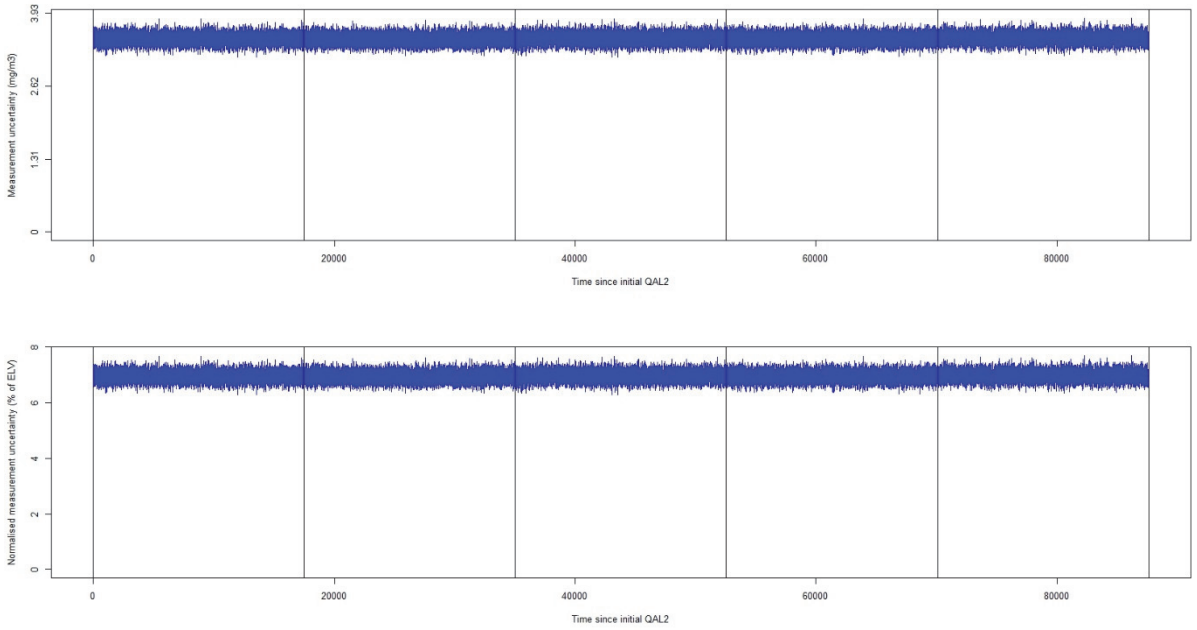

Figure 17 Concentration measurement uncertainty for each measurement after the initial QAL2 in mg/m^3 (top) and as a percentage of the ELV (bottom). Vertical lines indicate end of year AST testing. Plot is from a run with response to three interfering species with opposing tendencies set to 10%

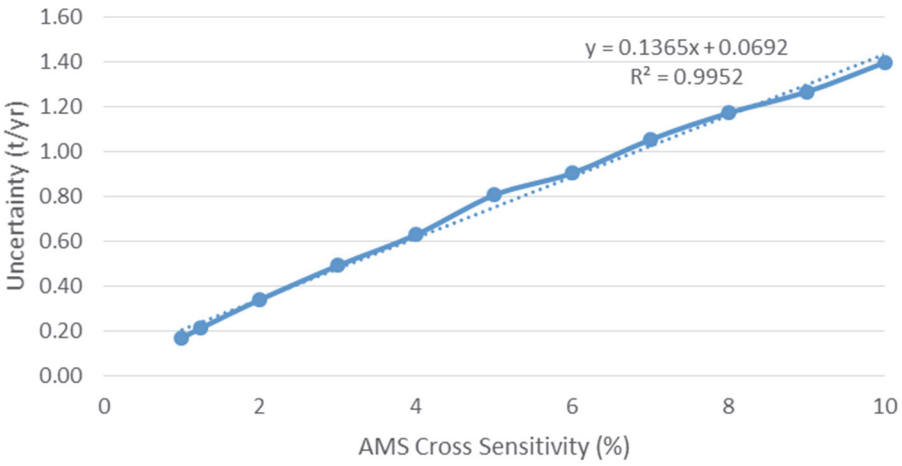

Figure 18 Overall uncertainty in annual mass emissions at varying levels of AMS cross sensitivity to three species with conflicting effects

3.2.5 AMS detection limit

Table 8 Model results indicating overall uncertainties when varying the AMS detection limit characteristic

AMS detection limit (%)	Concentration overall uncertainty (mg/30 minutes)	Highest recorded concentration uncertainty (mg/30 minutes)	Mass emission overall uncertainty (t/year)
1.5	0.2549	0.2962	0.2124
1	0.1629	0.1868	0.1372
2	0.3280	0.3813	0.2681
3	0.5009	0.5779	0.4204
4	0.6468	0.7469	0.5302
5	0.8170	0.9338	0.6488
6	0.9827	1.1182	0.8086
7	1.1388	1.2970	0.9326
8	1.2979	1.4951	1.0832
9	1.4291	1.6230	1.1884
10	1.5407	1.7886	1.2279

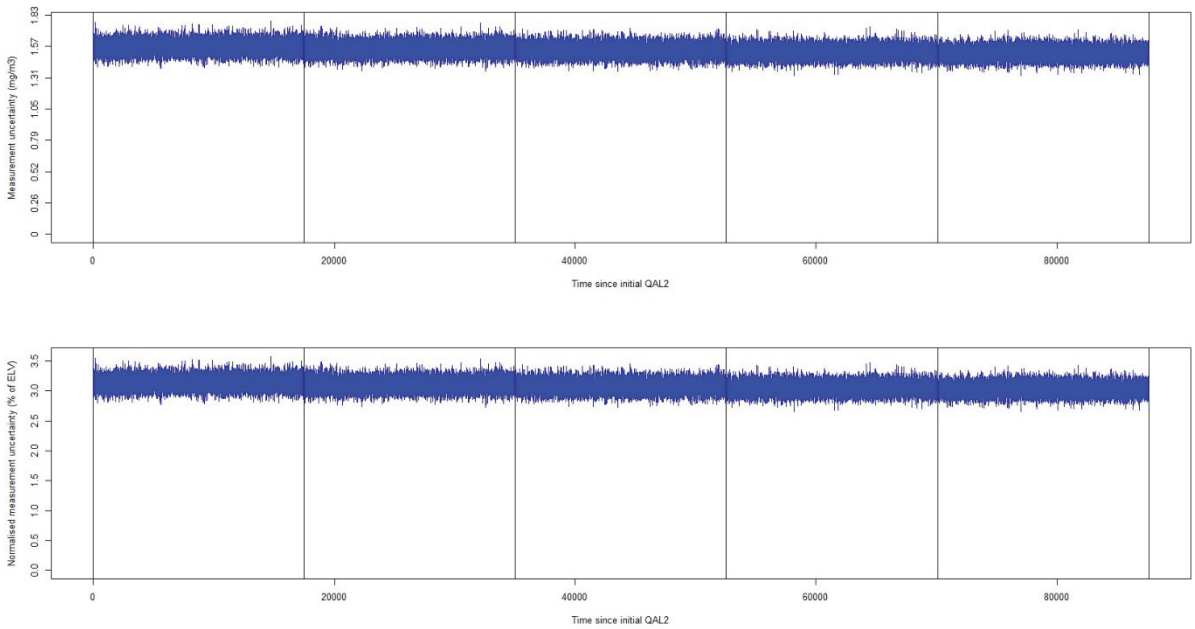

Figure 19 Concentration measurement uncertainty for each measurement after the initial QAL2 in mg/m^3 (top) and as a percentage of the ELV (bottom). Vertical lines indicate end of year AST testing. Plot is from a run with AMS detection limit set to 10%

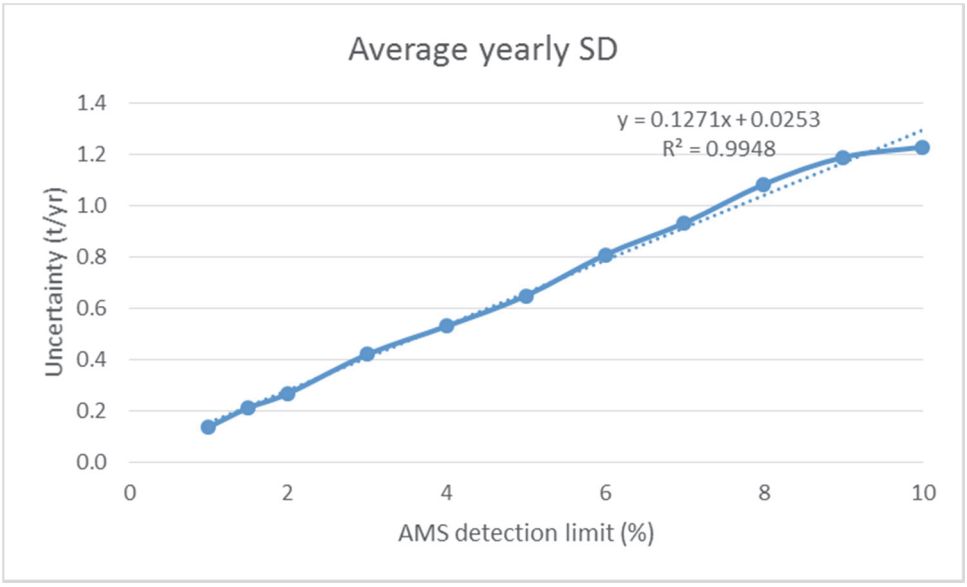

Figure 20 Overall uncertainty in annual mass emissions at varying AMS detection limit values

3.2.6 AMS measurement temperature offset from calibration temperature

Two sets of results as the temperature offset between calibration and measurement changes at different rates at zero and span levels. These have been tested separately with each varied while the other was set to zero. The initial calibration temperature is 15°C and the model uses a sine curve for the measurement temperatures to represent seasonal variations. An additional sine curve can be added to this to generate diurnal temperature variations but is not used in these tests.

Table 9 Model results indicating overall uncertainties when varying the AMS zero sensitivity to offset between measurement and calibration temperatures

AMS temperature offset - zero (%)	Concentration overall uncertainty (mg/30 minutes)	Highest recorded concentration uncertainty (mg/30 minutes)	Mass emission overall uncertainty (t/year)
1.58	2.0582	3.2201	2.2745
1	1.3487	2.0240	1.5481
2	2.6136	3.9612	2.9768
3	4.2094	6.2751	4.7095
4	5.5803	8.4420	6.1816
5	7.0509	10.4388	7.9303
6	8.8303	13.6541	9.9209
7	10.3981	16.8363	11.8276
8	11.7480	18.0762	12.8606
9	12.8013	19.8578	14.9410
10	14.8389	24.0562	16.5658

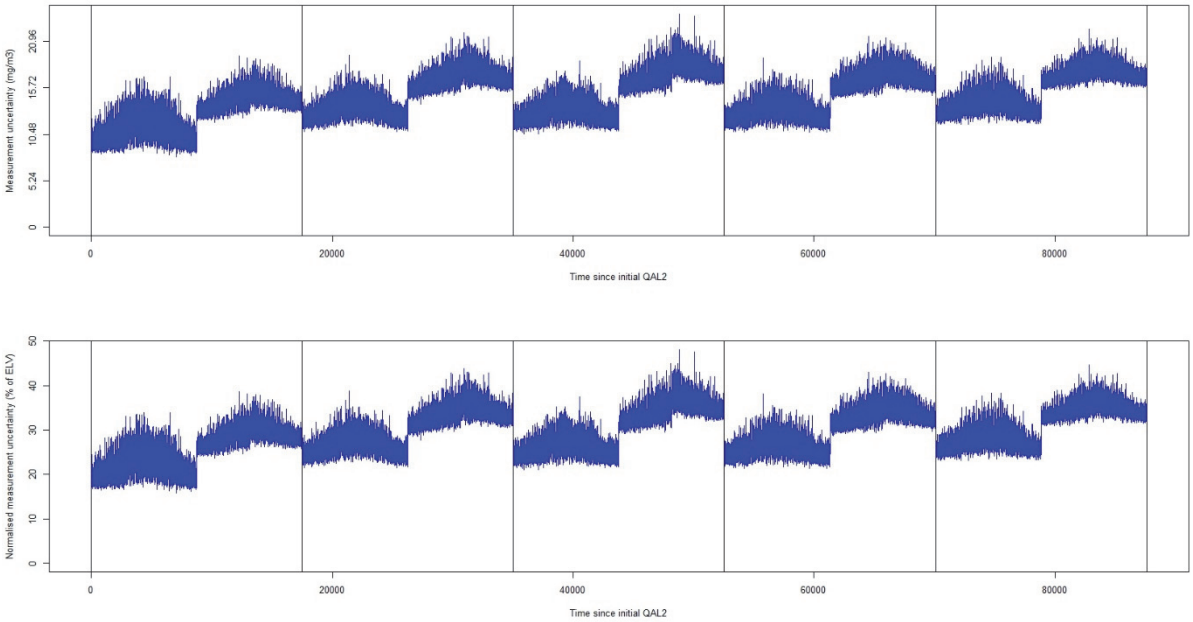

Figure 21 Concentration measurement uncertainty for each measurement after the initial QAL2 in mg/m^3 (top) and as a percentage of the ELV (bottom). Vertical lines indicate end of year AST testing. Plot is from a run with AMS temperature sensitivity at zero set to 10%

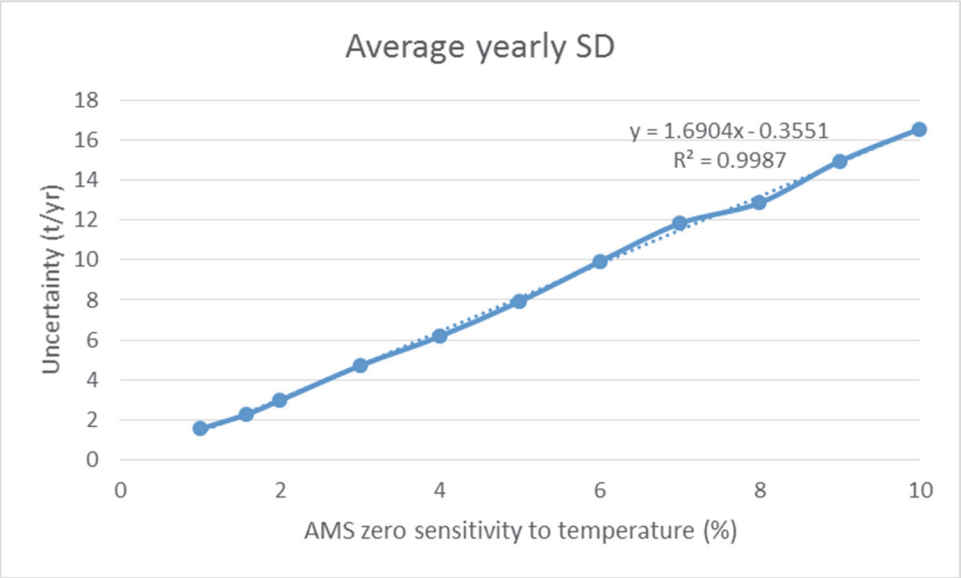

Figure 22 Overall uncertainty in annual mass emissions at varying levels of AMS zero sensitivity to offset between measurement and calibration temperatures

Table 10 Model results indicating overall uncertainties when varying the AMS span sensitivity to offset between measurement and calibration temperatures

AMS temperature offset - span (%)	Concentration overall uncertainty (mg/30 minutes)	Highest recorded concentration uncertainty (mg/30 minutes)	Mass emission overall uncertainty (t/year)
0.04	0.0277	0.0465	0.0268
1	0.7529	1.1905	0.8069
2	1.5195	2.2590	1.6319
3	2.4090	3.7712	2.6414
4	4.1770	9.8765	5.3862
5	5.2870	9.5730	6.8022
6	9.9085	39.6057	14.0476
7	13.6566	49.0792	19.3821
8	15.2091	55.5670	21.6319
9	18.0017	57.1420	25.5862
10	19.9967	49.1811	28.1001

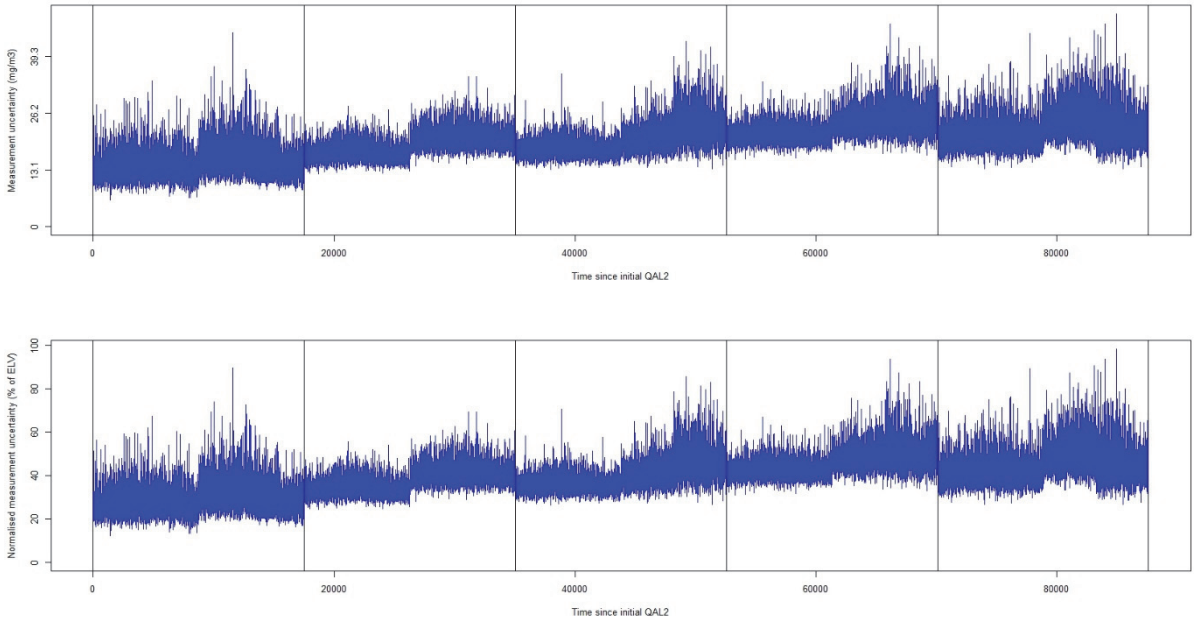

Figure 23 Concentration measurement uncertainty for each measurement after the initial QAL2 in mg/m^3 (top) and as a percentage of the ELV (bottom). Vertical lines indicate end of year AST testing. Plot is from a run with AMS temperature sensitivity at span set to 10%

3.2.7 AMS drift

Two sets of results as the instrumental drift changes at different rates at zero and span concentrations. These have been tested separately with each varied while the other was set to zero. The drift rates are the amount the instrument is expected to drift by up to between QAL3 tests.

Table 11 Model results indicating overall uncertainties when varying the drift rate on the AMS zero

AMS drift - zero (%)	Concentration overall uncertainty (mg/30 minutes)	Highest recorded concentration uncertainty (mg/30 minutes)	Mass emission overall uncertainty (t/year)
1.1	1.8120	2.8563	2.8172
1	1.7196	2.6433	2.6744
2	2.3895	3.4704	3.6900
3	2.6166	4.0415	3.9632
4	2.9090	4.7610	4.3040
5	3.2599	5.5055	4.7336
6	3.5712	6.1148	5.0805
7	3.6450	6.1339	5.1313
8	3.9899	6.6704	5.6111
9	4.1853	6.9719	5.8306
10	4.5888	7.6317	6.3778

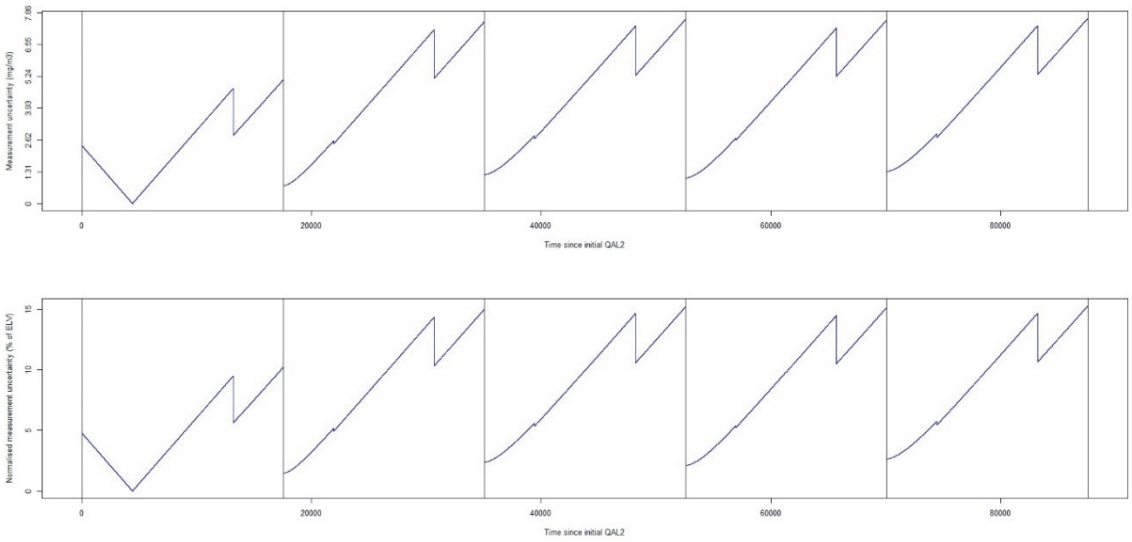

Figure 24 Concentration measurement uncertainty for each measurement after the initial QAL2 in mg/m^3 (top) and as a percentage of the ELV (bottom). Vertical lines indicate end of year AST testing. Plot is from a run with AMS zero drift set to 10%

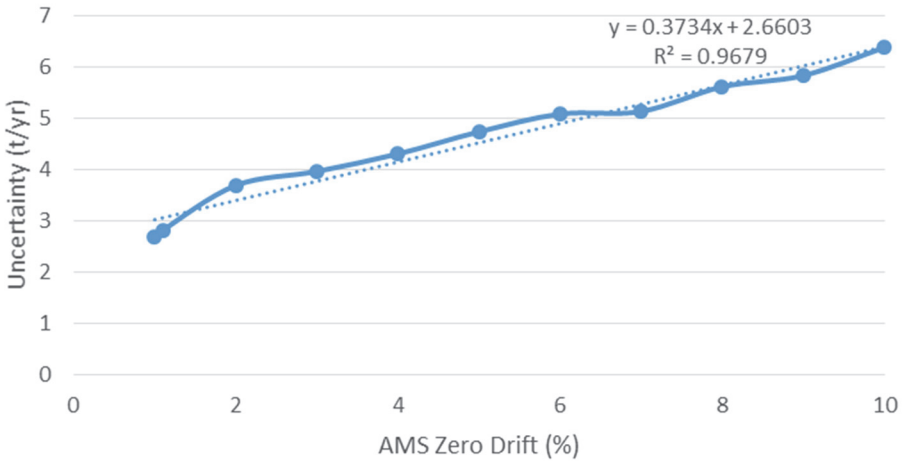

Figure 25 Overall uncertainty in annual mass emissions at varying levels of AMS zero drift

Table 12 Model results indicating overall uncertainties when varying the drift rate on the AMS span

AMS drift - span (%)	Concentration overall uncertainty (mg/30 minutes)	Highest recorded concentration uncertainty (mg/30 minutes)	Mass emission overall uncertainty (t/year)
1.9	1.4661	2.1267	2.2713
1	1.1091	1.7375	1.7300
2	1.5005	2.1802	2.3195
3	1.6254	2.6210	2.4573
4	1.8068	3.0641	2.6646
5	2.0534	3.4807	2.9779
6	2.1782	3.7413	3.1040
7	2.2914	3.9359	3.2266
8	2.4862	4.2324	3.4811
9	2.6449	4.5135	3.6581
10	2.8937	4.9104	4.0012

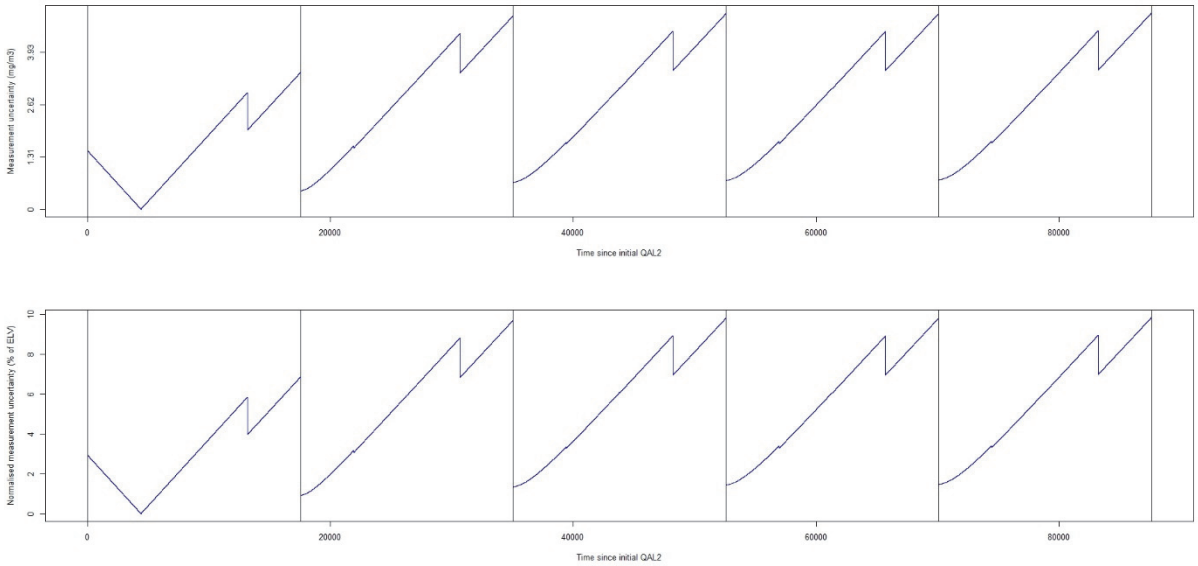

Figure 26 Concentration measurement uncertainty for each measurement after the initial QAL2 in mg/m^3 (top) and as a percentage of the ELV (bottom). Vertical lines indicate end of year AST testing. Plot is from a run with AMS span drift set to 10%

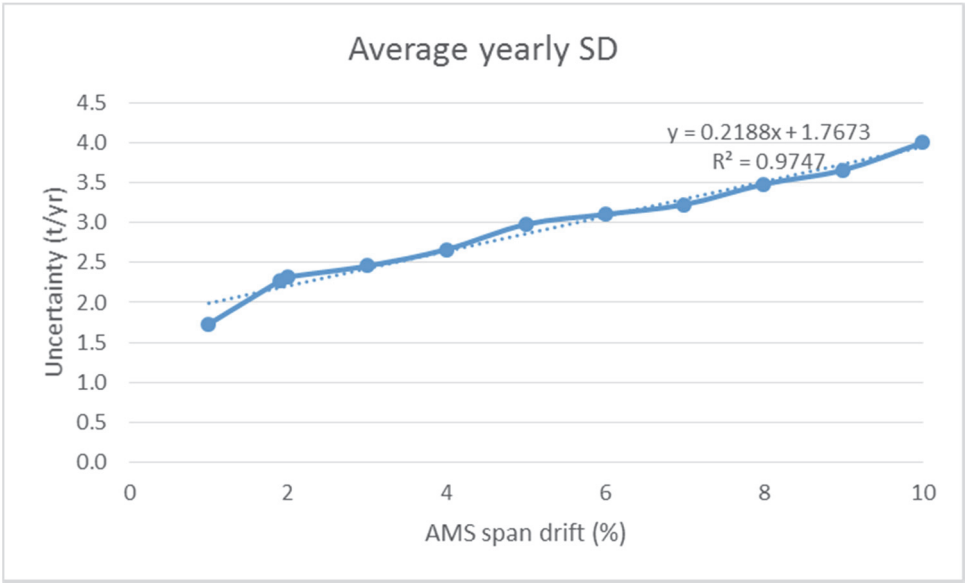

Figure 27 Overall uncertainty in annual mass emissions at varying levels of AMS span drift

3.2.8 AMS O_2 uncertainty

Concentration values are typically reported at standard conditions, including oxygen levels. Correction for this requires oxygen levels in the flue to be monitored. These values are not "measured" in the model and the correction is not applied to measurements except for quality assurance testing. Currently this does not influence the overall uncertainty produced by the model.

3.2.9 AMS H_2O uncertainty

Concentration values are typically reported at standard conditions, requiring conversion from a wet gas flow so moisture levels have to be monitored. These values are not "measured" in the model and the correction is not applied to individual measurements. Moisture correction is only applied to the

quality assurance testing procedures, so the uncertainty associated with it does not contribute to the overall uncertainty in the model.

3.2.10 Sensitivity summary

With the concentration and flow rates used in all the model runs the annual mass emissions should be 39.42 tons per year. The mass emission uncertainties have been converted to a percentage of that value so they are directly comparable (Table 13).

Table 13 Model attributes and their impact on overall uncertainty when set to 10%

Attribute	Overall concentration uncertainty with attribute uncertainty at 10% (mg/30 minutes)	Overall mass emission uncertainty with attribute uncertainty at 10% (t/year)	Mass emission uncertainty as a percentage of the actual mass emissions (%)
SRM uncertainty	0.659	1.039	2.64
AMS repeatability	3.368	3.687	9.35
AMS linearity	2.739	2.199	5.58
Cross interference: 1 species	2.237	0.881	2.24
Cross interference: 3 species - complementary	3.460	1.364	3.46
Cross interference: 3 species - conflicting	3.472	1.396	3.54
AMS detection limit	1.541	1.228	3.11
AMS temperature offset from calibration - zero	24.056	16.566	42.02
AMS temperature offset from calibration - span	19.997	28.100	71.28
AMS zero drift	4.589	6.378	16.18
AMS span drift	2.894	4.001	10.15

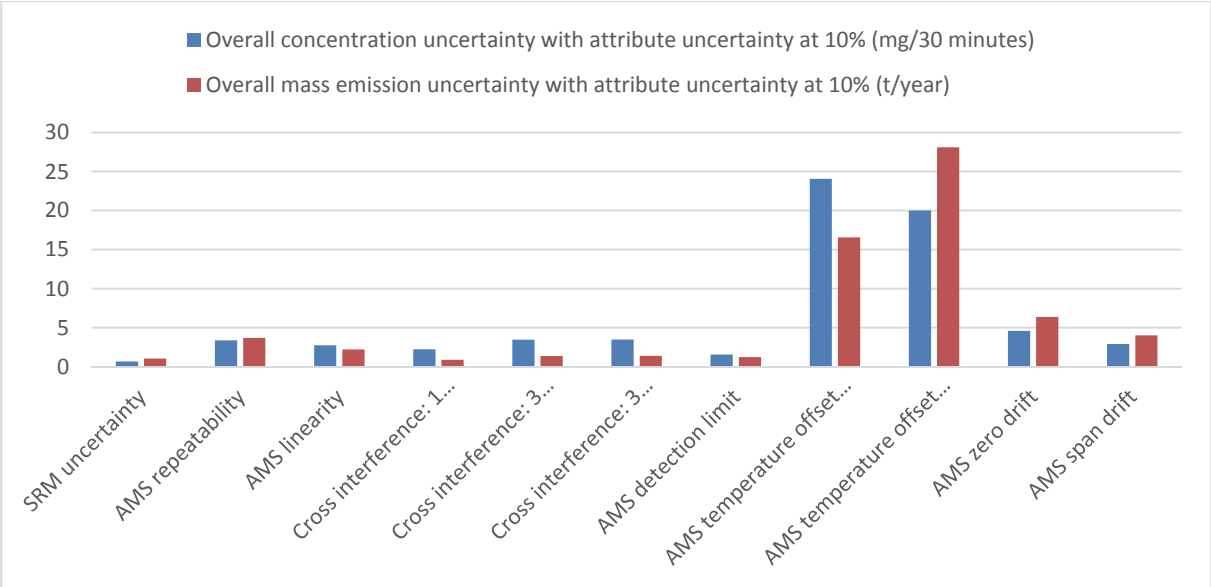

Figure 28 Sensitivity of concentration and overall mass emission uncertainty to the modelled attributes set at 10%

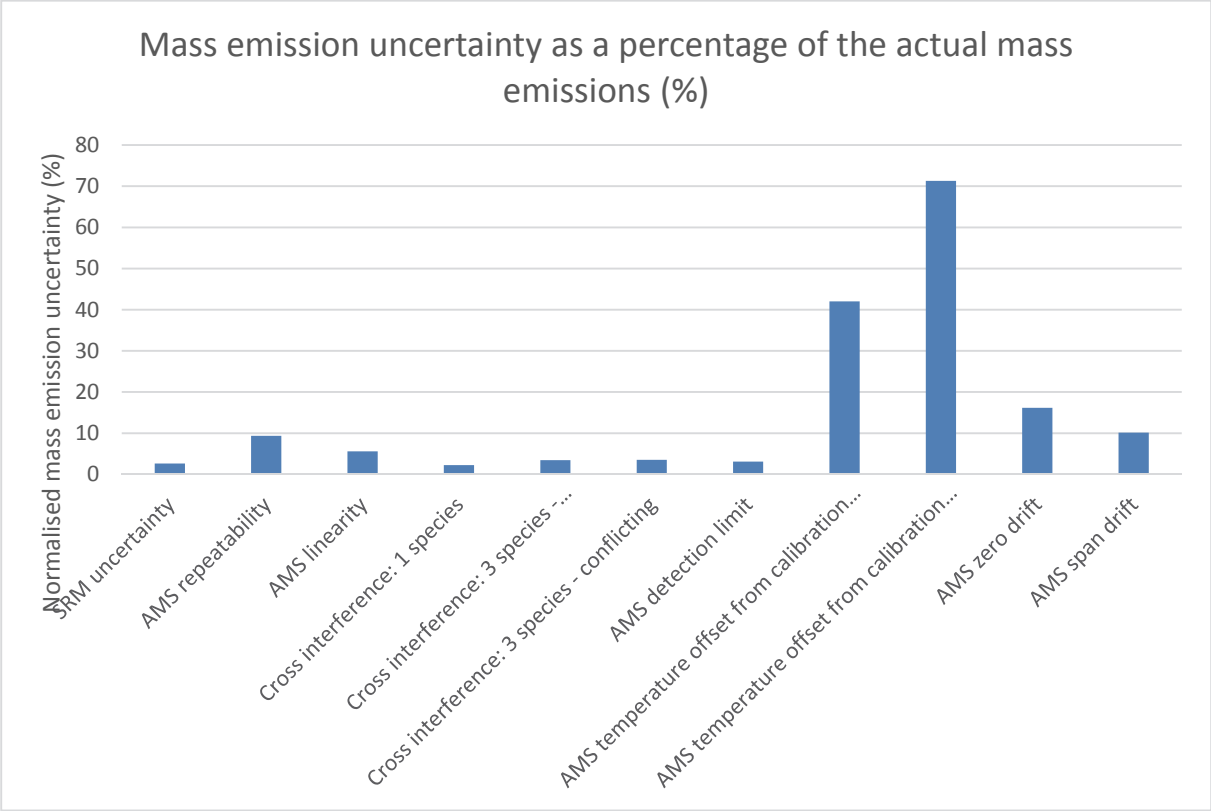

Figure 29 Sensitivity of overall mass emission uncertainty as a percentage of the actual emissions to the modelled attributes

4 CONCLUSIONS AND DISCUSSION

The sensitivity testing of the full model supported the results of the validation testing using real data. When SRM uncertainty moves above 15% there is a marked increase in AST failures due to the poor quality SRM. The sensitivity testing showed that the growth in overall uncertainty stayed linear even when SRM uncertainty moved above 20%, demonstrating that having a limit at that point makes very

little difference. Lowering the limit on SRM uncertainty to ±15% would reduce potential difficulties with good quality CEMs failing due to poor SRM performance. Improvements to overall uncertainty would be minor, but the real benefit would be fewer failed ASTs and the subsequent additional QAL2s, something that will be welcomed by operators.

The AMS repeatability demonstrated a reduction over time in the overall uncertainty. The maximum uncertainty of any measurement (Figure 8) continues to increase as the AMS repeatability uncertainty rises, but over time a mix of QAL3 and AST failures leads to recalibrations that improve the overall performance in subsequent years (Figure 7). When there is a failure the model runs a fresh QAL2 on that repeat. This too may generate a poor calibration function, causing another failure, but if this goes on long enough the technique will self-select for better instruments, improving the overall uncertainty. This could be viewed as a flaw in the model as it gives an incomplete indication of the situation in later years, but this is an accurate representation of the reality as poor instruments are likely to be identified and either fixed or replaced if there are repeated failures.

The AMS linearity uncertainty leads to cumulatively larger overall uncertainty, with relatively high numbers of QAL3 failures between ASTs causing discontinuities in the concentration measurement uncertainty (Figure 10). Interestingly while the resets due to failing QAL3 tests tend to make overall uncertainty worse, resets performed due to failed ASTs appear to reduce the overall uncertainty. This would indicate that the mode of failure might have an influence on the effects of self-selection. The QAL3 control charts are based on the s_{AMS} value, so it may be that this should be altered to help prevent repeats with "better" calibrations failing the QAL3.

For cross interfering species, where the magnitude of the related errors is affected by the effectiveness and concentration of one or more conflicting species, the effects on overall uncertainty varied according to the number of species and their influence (positive or negative) on the measured value. Extra interfering species increased the overall uncertainty, although the direction of the influence had little impact on the magnitude of this effect, as the tests with three species partly cancelling each other out caused similar overall uncertainty to runs where all three species generated complimentary errors, with both achieving a mass emission uncertainty ~3.5% when set to 10% characteristic uncertainty.

Higher detection limits lead to higher overall uncertainty, although the sensitivity of this attribute is lower than many others. With a mass emission uncertainty of 3.1%, only SRM uncertainty and cross interference with a single species had less effect on overall uncertainty at 10% intensity than AMS detection limits.

The use of quarterly QAL3 testing caused the model to encounter step changes in temperature between tests, resulting in failures even at low temperature offset uncertainties. More frequent QAL3 testing would prevent this as temperature in the model only changes gradually which might prevent triggering the failures dependant on the s_{AMS} values being used. The more frequent recalibrations will have affected the modelled overall uncertainty for temperature effects so the overall uncertainty of 42% for zero and 71% for span might not be directly comparable with other factors.

The temperature offset span effect led to some extreme calibration functions in the model (b-values of more than a hundred). In order to prevent this from skewing results a check was put in on the calibration function to ensure that unrealistic calibration functions were not accepted. Even with this sanity test in place the temperature offset span effect was the factor with the highest sensitivity (71%). The use of a check on extreme calibration functions is justified as this is part of the guidance for Danish QAL2 testing (FORCE Technology, 2015). The limits specified in the guidance are for b to be close to one (±0.2), however the constraints in the model were relatively weak, allowing b values ±20 since additional constraints could block natural behaviour that should be encapsulated in the model.

The high sensitivity of the overall uncertainty to the ambient temperature demonstrates the need to calibrate instruments at representative conditions to keep offsets low. The model was simulating

offsets of up to 15°C in the worst cases. Where possible instruments should be kept in temperature controlled environments to reduce the influence of ambient temperatures.

At low drift rates the QAL3 adjustments are successful in restricting measurement uncertainty. In later years of a run the model indicates a potential for some failed ASTs, but this ensures the AMS operates within the required uncertainty limits. As drift rates increase the periodicity of the QAL3 tests becomes a problem and the adjustments are not able to keep up. Failures maintain uncertainty levels, but more frequent QAL3 testing would likely prevent this from becoming an issue until much higher drift rates.

5 FURTHER WORK

The model has been designed in order to allow the modification of many of the variables and influence factors, so there are plenty of opportunities to investigate the effects of alternative changes on overall uncertainty of emissions monitoring.

One area that operators are most vocal about is the frequency of testing, in particular for the routine QAL3 for drift detection. Testing often takes instruments offline and operators have to meet reliability requirements. However the primary concern is the cost, both financially and in employee time required for carrying out the testing regime. NPL plans to look into possibilities for altering testing frequency and what effect that would have on meeting the uncertainty limits. The model was designed to be flexible, allowing the timings of QAL3 and AST testing to be altered to see the effects on overall uncertainty.

Additionally work on the model is ongoing, adding features such as adjustable diurnal temperature variation. Flow within the model is somewhat simplistic at this point and could be improved in future, incorporating influence factors for swirl and to indicate the effect of in homogeneity in the flow profile. Adding conversion to standard conditions (oxygen and moisture correction) for all measurements in the model would improve the quality of results as currently this only applies to the quality assurance procedures and does not contribute to the overall uncertainty.

The model currently works on half hourly average measurements, so one potential future direction would be to take the scope down to individual measurements. This would show the effects of averaging to get the reported half hourly concentration and flow measurements. At this scale you would have to include factors relating to misalignment if measurements from different sources are not temporally aligned. This could occur due to differences in clock settings or if sample lines for different instruments have varying transit times, etc.

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7 ANNEX 1: SUMMARY OF MODEL VARIABLES WITH DESCRIPTION OF TYPE AND CONTROL MECHANISM

Variable	Type	Summary
True measured gas concentrations	Values	Input argument
Number of measurements between QAL2	Constant	Set in parameter file
Number of measurements between QAL3	Constant	Set in parameter file
AMS linearity error	Uncertainty	Set in parameter file
AMS species 1 cross sensitivity error	Uncertainty	Set in parameter file
AMS species 2 cross sensitivity error	Uncertainty	Set in parameter file
AMS species 3 cross sensitivity error	Uncertainty	Set in parameter file
Maximum calibration limit	Constant	Set in parameter file
AMS detection limit	Uncertainty	Set in parameter file
Emission limit	Constant	Set in parameter file
AMS repeatability	Uncertainty	Set in parameter file
AMS calibration temperature	Values	Set in parameter file
AMS temperature zero drift	Uncertainty	Set in parameter file
AMS temperature span drift	Uncertainty	Set in parameter file
AMS time zero drift	Uncertainty	Set in parameter file
AMS time span drift	Uncertainty	Set in parameter file
AMS uncertainty from instability	Uncertainty	Set in parameter file
AMS uncertainty from variations in ambient temperature	Uncertainty	Set in parameter file
AMS uncertainty from variations in voltage	Uncertainty	Set in parameter file
AMS uncertainty from variations in ambient pressure	Uncertainty	Set in parameter file
AMS uncertainty from any other influences	Uncertainty	Set in parameter file
SRM overall uncertainty	Uncertainty	Set in SRM parameter file
Measurement temperatures	Values	Yearly files of measurement temperatures
Number of parallel measurements for QAL2	Constant	Function input argument (default=15)
Uncertainty of O2 measurements	Uncertainty	Set in parameter file
Uncertainty of H2O measurements	Uncertainty	Set in parameter file
Uncertainty of cross-sectional area of the duct	Uncertainty	Set in parameter file
Uncertainty in the flow rate measurement	Uncertainty	Set in parameter file