Publishable Summary for 16ENV08 IMPRESS 2
Metrology for air pollutant emissions

Overview

European Directives came into force setting increasingly stringent Emission Limit Values (ELVs) for key air pollutants to mitigate ~400 000 premature deaths and €330-€949 billion p.a. of costs attributable to air pollution. However, the emission monitoring industry faced an issue, as a metrologically robust framework of standardised measurement methods to enforce these directives was not fully in place. This project addressed this measurement gap by innovating measurement methods for new measurands (NH_3, formaldehyde, HF, PM, SVOC, OGC, PAH), addressed a lack of uncertainty characterisation in flow measurements and developed next generation techniques for increasingly stringent future legislation. These improvements and innovations in measurement and uncertainty assessment provide the capability to enforce further tightening of ELVs in future.

Need

Emission limits are enforced by measurements using techniques (instruments) operated in accordance with documentary standards published by CEN, these conventionally being referred to as Standard Reference Methods (SRMs) and either being passed into, or referred to, in member state legislation. There are now European directives regulating emissions from large scale processes, such as power stations, all the way down to domestic boilers burning fuels such as wood pellets. New directive requirements have a two-fold impact in that they are bringing in ELVs for previously unregulated emissions species and also increasingly stringent ELVs for species currently regulated. This exposed gaps in the capabilities of techniques and SRMs, potentially undermining the ability of national regulators for fulfilling their legal responsibilities to enforce such limits. On the large scale there were no SRMs for HF, NH_3 and formaldehyde, and there were questions over whether the existing SRMs for HCl and dust would be able to enforce increasingly stringent ELVs. With respect to small-scale biomass, there were no SRMs for semi-volatile organic compounds (SVOCs), organic gaseous carbon (OGC), polyaromatic hydrocarbons (PAHs) or particulate matter (PM). There was no on-line measurement technique to apportion CO_2 emissions as renewable or fossil fuel derived. Also, cutting across all industrial processes there were some significant issues with flow uncertainty, as with small ducts there was a lack of any work identifying and quantifying sources (needed for dust measurements) and with large processes work was needed to validate existing uncertainty knowledge and develop novel lower uncertainty techniques. Lastly, as acknowledged by the EC, the current legislation would not meet WHO air quality guidelines, and so work was needed on the next generation of techniques to enforce ELVs in future legislation.

Objectives

The overall aim of this project was to provide metrology to enable the enforcement of the Industrial Emissions, Medium Combustion Plant and Eco-design Directives, and the EU's Emissions Trading Scheme.

The scientific and technical objectives of the project were as follows:

1. To develop validated reference measurement methods where currently none exist for HF, NH_3, formaldehyde and to test the limitations of the existing HCl SRM (EN 1911) and dust SRM (EN 13284-1) for enforcing increasingly stringent ELVs**.** To develop next generation optical techniques laying the platform to enforce ELVs in future legislation.
2. To develop validated reference measurements methods for SVOCs, OGC, PAHs (including benzo[a]pyrene) and PM from small scale combustion sources meeting Eco-design directive uncertainty requirements.

The EMPIR initiative is co-funded by the European Union's Horizon 2020 research and innovation programme and the EMPIR Participating States

3. To develop hyperspectral multispecies methods for the determination of emissions from biomass combustion including non-wood fuelled. To develop an on-line, real-time technique to apportion CO_2 emissions between renewable and fossil fuel in co-firing biomass plants.
4. To determine the uncertainty and traceability of mass emission measurement with respect to flow calibrations under field conditions. To establish the impacts of wall effects and sensor obstruction, particularly in small ducts. To investigate the use of multiple sensors in stacks and the potential to decrease flow uncertainties
5. To provide input to the development and/or revision of standards related to the emissions of semi-volatile organic compounds (SVOCs), particulate matter (PM), polyaromatic hydrocarbons (PAHs) and organic gaseous carbon (OGC). In addition, to facilitate the take up of the technology and measurement infrastructure developed in the project by standards developing organisations (such as CEN TC 264 and ISO TC 146 and those linked to the EU Eco-design Directive 2009/125/EC, MCP Directive 2015/2193 and IED 2010/75/EU) and end users (e.g. environmental monitoring and regulation bodies, the power generation sector, combustion equipment manufacturers).

Progress beyond the state of the art

Project partners developing and validating protocols for measuring unregulated measurands (HF, NH_3 and formaldehyde) or new more accurate methods for measuring existing regulated emissions (HCl and dust), opens the door to a step change in future regulation. Lower emission limits will lead to better air quality and a subsequent reduction in excess deaths related to industrial emissions.

Improvements in large scale industrial emissions has led to an increasing proportion of emissions to come from smaller scale biomass burning. The project has contributed new measurement methods for SVOCs, OGC, PAHs (including benzo[a]pyrene) and PM, including the development of a dilutor sampling approach for this monitoring. Residential biomass burning is especially important to regulate as it occurs directly within population centres and this project has helped develop the measurement techniques and protocols required for future monitoring.

The project has developed novel hyperspectral, multispecies capability to monitor emissions from both biomass and other combustion sources. The real-time, on-line instrument to apportion CO_2 emissions from co-firing power stations was field tested as part of the project validating its potential to enable a shift from fossil fuels to biomass, reducing the carbon intensity of the power sector.

Most previous efforts have focused on the concentration measurement of emissions so that is relatively well quantified. This project has investigated the uncertainties in flow measurement that is combined with concentration data to report mass emissions of pollutants. With the introduction of the MCP Directive monitoring flow in narrower ducts has become more common, so the knowledge gained in this project on blocking, wall effects, and turbulent flow will make a significant difference for future monitoring.

Results

Objective 1: To develop validated reference measurement methods where currently none exist for HF, NH_3, formaldehyde and to test the limitations of the existing HCl SRM (EN 1911) and dust SRM (EN 13284-1) for enforcing increasingly stringent ELVs. To develop next generation optical techniques laying the platform to enforce ELVs in future legislation.

The INERIS and NPL stack teams both took part in the proficiency testing schemes run by both organisations. NPL wrote a report comparing the schemes and analysis of the data demonstrated that each team had performed better in their home scheme. This indicates that participants are likely to do better when they are more familiar with a PT scheme.

An NPL facility to simulate dust measurement from a duct was used to investigate the ability of current measurement techniques to measure at low concentrations. The limit of detection for current methods is still capable of detecting dust at the required levels, but measurement uncertainty is being challenged by lower emission limit values.

INERIS and NPL reviewed the available techniques for measuring HF, NH_3 and formaldehyde, assessing their relative strengths and weaknesses, concluding that current methods will not be capable of the required measurements if the ELV is reduced.

By using two different spectroscopic techniques CNR verified, in a stack simulator, the effects of gas composition on the readings of gas analysers based on laser spectroscopy. Most commercial devices adopt a detection technique named "derivative spectroscopy", which must be carefully calibrated when the gas mixture under investigation features changing concentrations of water or CO_2, in particular when measuring polar gases, like ammonia or hydrogen chloride.

INERIS carried out an inter-comparison of manual methods for NH_3. The results demonstrated the equivalence of the reference method and Gasmet DX 4000 FTIR, however the required calibration function varied depending on the matrix of combustion gases, particularly when the emissions came from biomass combustion.

NPL produced procedures for in-field wet calibration for HCl and NH_3 measurements, along with a documented uncertainty budget. This highlighted that sample losses in measurement equipment can increase measurement uncertainty by 5 %.

PTB developed a direct tunable laser absorption spectroscopy (dTDLAS) for making low concentration HCl measurements, since the current reference method is unable to measure at the lowest levels likely to be required in future.

DTU carried out laboratory and field testing of a prototype optical instrument for measuring formaldehyde and NH_3. This included testing at the INERIS test bench which demonstrated high sensitivity for NH_3/H_2CO and agreement with INERIS data.

From a review of techniques and practical experiments INERIS was able to develop a set of protocols for HF, NH_3 and formaldehyde. This assesses the suitability of all the available techniques to see if they can meet the required uncertainty of ≤20 % of the applicable ELV. The development of next generation techniques by INERIS has the potential to meet this requirement in the case of further lowering of the emission limits in future.

A blind inter-comparison study investigating HCl reference method lab uncertainties was carried out by NPL. This showed that significant numbers of results would not meet the overall uncertainty requirement for current IED emission limits (21.6 %) or future BAT conclusion limits (54.9 %). This highlights the need for future revision of EN 1911 to focus on improving the uncertainty requirement for laboratory analysis. Monte-Carlo model analysis further supported this conclusion.

This objective was successfully achieved.

Objective 2: To develop validated reference measurements methods for SVOCs, OGC, PAHs (including benzo[a]pyrene) and PM from small scale combustion sources meeting Eco-design directive uncertainty requirements.

NPL carried out a laboratory inter-comparison by sending traceable solutions for PAH analysis to four accredited European laboratories. The spread of results from the different analytical laboratories calls into question the uncertainty currently stated for the method. Regular proficiency testing, as is required for other pollutants, would highlight issues at accredited laboratories for PAH analysis, leading to improved performance in future.

INERIS carried out an inter-laboratory comparison to assess the measurement performance of participants for CO, NO_x, TOC, dust, SO_2, HCl and HF. Using data from nine rounds of the ILC and some additional data produced for the project using the SRMs, INERIS and NPL have produced a paper to look at how the measurement uncertainty compares to requirements in current and future legislation. The AMS methods met the IED guidelines, but, focussing on the SRM measurements, the results demonstrated that the reference methods for CO, TOC and dust were already falling short of the IED requirements. For HCl (reaction with ammonia) and HF (too few results available) the results were inconclusive about the uncertainty requirement. Lower ELVs that have already been proposed in existing BAT conclusions will expand this issue, so new measurement methods, particularly new SRMs, are required to meet the upcoming measurement challenge.

DTU used UV absorption spectroscopy for OGC and PM measurements at the same time in a gas cell and identified issues with bias caused by soot deposition. This issue prevented in situ (cross-stack) measurement.

The Eco-design Directive covers emissions from biomass burners for domestic heating, however there are no European standards for measuring PM emissions for this application. RISE assessed the technical requirements for PM and SVOC monitoring, creating a measurement specification which was subsequently utilised in the formulation of protocols for future standardisation.

There are no standard reference methods for measuring SVOC, PAHs or condensable PM generated from small scale biomass like residential wood combustion. INERIS, RISE, ENEA, ISSI and DTI worked together to develop and validate three protocols to characterise these sorts of emissions.

- Using a dilution chamber to simply characterise solid and condensable fraction of particulate matter
- Simplified method of characterisation for PAH emissions including BaP, a regulated compound in ambient air which is mainly generated by residential combustion
- Method for characterisation of SVOC emissions according to volatility characterisation

Using these methods would improve emission inventories and close the gap between observed and modelled PM in ambient air. Existing standards are not sufficient to characterise these, so the developed protocols are being put forward for adoption as new standards under CEN TC295.

This objective was successfully achieved.

Objective 3: To develop hyperspectral multispecies methods for the determination of emissions from biomass combustion including non-wood fuelled. To develop an on-line, real-time technique to apportion CO2 emissions between renewable and fossil fuel in co-firing biomass plants.

As biofuels begin to replace fossil fuels it is important to be able to validate the relative proportions being combusted in co-firing plants. This can be achieved by investigating the isotopic ratio of CO_2 in the emissions. To achieve the required sensitivity, it is necessary to use a dilution system and remove potential interferents. VTT developed an isotope analyser and validated it in both laboratory and field experiments. The results provide the potential to use online spectroscopic methods for apportion of CO_2 emissions between renewable and fossil fuels.

CEM, UC3M and VSL carried out a survey of current technology for hyperspectral detection of emissions. VSL followed on by developing an instrument for measuring HCl from biomass emissions, but this requires further development. CEM and UC3M developed a calibration and validation procedure for remote quantification based on reference gases produced and certified by CEM. UC3M have achieved excellent results using the technique during the validation testing at the UC3M and VSL facilities.

This objective was successfully achieved.

Objective 4: To determine the uncertainty and traceability of mass emission measurement with respect to flow calibrations under field conditions. To establish the impacts of wall effects and sensor obstruction, particularly in small ducts. To investigate the use of multiple sensors in stacks and the potential to decrease flow uncertainties.

The NPL Monte-Carlo model was also used to investigate the propagation of uncertainties in the dust and flow reference methods into reported annual mass emissions. Evaporation losses were the most significant source of error for the dust reference method, followed by leak rate and misalignment of the sampling nozzle. For flow there are multiple reference methods that can be used, so the NPL Monte-Carlo model was used to compare uncertainties from L-type Pitot, S-type Pitot, vane anemometer and hot-wire anemometer. The model showed Pitot tubes were the best reference method, although there was a tendency for all modelled techniques to calibrate a small negative bias that would result in systematic under reporting of emissions. Measurement standards assume bias free calibration, so further development to improve techniques is required to meet this requirement.

Optical techniques proved to be simple and reliable tools for the calibration of point flow sensors (e.g. Pitot probes) in narrow ducts. With stack diameters lower than 0.35 m, any small perturbation of the flow can produce local turbulence, negatively affecting the readings of standard probes. This effect can be strongly enhanced by disalignments of the probe axis with respect to duct axis. A measurement tool, based on spectroscopic techniques, was developed within this project, suitable for fast deployment in real environments.

Two different sources of error of flow rate measurement in stacks have been investigated assuming the flow rate is measured according to the Standard Reference Method (EN ISO 16911-1) using pitot tubes in a grid of points. The error due to a pitot tube sensitivity to inclined flow direction and the error due to number and position of the sampling points have been discussed using the measured data provided by VSL and numerical simulation data from CMI.

It was found both types of pitot tube have comparable sensitivity to the flow angle. The errors are less than 2 % for distance from a T-junction larger than 3D. The minimal requirements of the SRM of having one sampling point for narrow ducts and five or more hydraulic diameters straight duct upstream from the sampling plane, do not exclude potential errors around 10 % and 15 % for straight and elbowed supply pipe, respectively.

Based on the results, it can be concluded that both the number and location of sampling points need an appropriate assessment, also considering the shape of supply pipe since changes immediately before the monitoring site can lead to unstable turbulent flow regimes which are challenging to monitor.

Supporting experimental work carried out by VSL, TU Delft used computational fluid dynamics modelling to investigate the effects of blocking and sampling close to the duct wall on flow measurements made by Pitot tubes. These can both become significant if the measurements are made in very small ducts, which is more likely at the smaller facilities covered by the medium combustion plant directive. Results indicate that as long as the minimum requirements in EN 16911, the standard for flow measurement in ducts, the measurements should be free of wall effects and blocking.

This objective was successfully achieved.

Impact

Impact on industrial and other user communities

NPL hosted an online stakeholder workshop to engage with industrial and regulatory stakeholders. This was well attended with more than 40 attendees and gave the opportunity to communicate the findings of the whole project directly to the important stakeholders, ensuring maximum impact.

VTT developed an instrument and sampling setup to apportion emission ratios of biomass and fossil fuels from CO_2 isotope analysis. This will enable combustion plants co-firing biomass and fossil fuels to report carbon emissions based on measurement of actual emissions rather than calculated based on fuel usage monitoring, which should lead to improvements in local, regional and national emission inventories. Reducing monitoring overheads in this way could encourage increasing use of co-firing, reducing the fossil fuel proportion in the fuel mix used at such facilities.

Achievements from this project are likely to lead to new reference methods allowing the setting of emission limits for previously unregulated measurands and the tightening of existing limits where the project has improved uncertainties enough to make lower limits enforceable. This will drive investment in new analytical instrumentation and associated sampling equipment. If the technologies developed in this project can demonstrate the ability to enforce lower emission limits, then it will be very difficult for policy makers to avoid casting future regulation to meet the 2050 target of meeting WHO air quality pollution guidelines. This compliance demand will result in further acquisitions of stack testing instrumentation increasing sales.

Impact on the metrology and scientific communities

The work in this project has highlighted the need for advances in measurement techniques for several measurands in order to meet the measurement challenge of lower emission limit values. Project partners have been developing and validating new techniques and protocols to standardise the measurements to meet this target. This is clearly a good problem to have as it indicates that process operators are lowering their emissions, leading to better air quality and a reduction in negative health effects on the general population.

The project has focused on field measurement issues including field validation of flow modelling work, testing NH_3 and formaldehyde measurement protocols on stack simulator facilities, investigating the limitations of the dust SRM and developing and testing the dilution chamber approach for residential scale biomass emissions sampling. This direct knowledge of real-world measurement challenges allows the metrology community to develop better instruments and methods for future use in the field.

The peer-reviewed papers that have come out of the project have significantly advanced the evidence base in the scientific literature in this field, especially since they are open access and available to the community. In the past, testing of alternative methods that were not approved would not be recorded. Failing to record these unsuccessful test campaigns leads to the reasons for rejection being lost, which could potentially lead to wasted effort repeating these tests. This evidence will benefit the community going forward.

Impact on relevant standards

Work by NPL and INERIS contributed to a draft standard for the determination of formaldehyde in ducts and stacks which is under preparation by WG40 of CEN/TC 264. Additionally, the protocols developed for HF and NH_3 have also fed into CEN/TC 264/WG 3 and ISO/TC 146/SC 1/WG 33 respectively.

INERIS, RISE, ENEA, ISSI and DTI developed protocols for measuring SVOC, PAHs or condensable PM generated from small scale biomass. These protocols are now being put forward for adoption as new standards under CEN/TC 295.

Longer-term economic, social and environmental impacts

Better monitoring techniques with lower uncertainties allow lower emission limits to be set and enforced, leading to improved air quality, which in turn is beneficial for human health and the environment. The European Commission stated that successful implementation of the Industrial Emission Directive would lead to a reduction in premature deaths / years of life lost in Europe of 13000 and 125000 respectively. This would save the EU €7 – 28 billion per annum.

The Clean Air Policy Package for Europe, which includes the Medium Combustion Plant Directive, is expected to avoid 58000 premature deaths per annum by 2030. This package is expected to save €40-140 billion per annum, compared with an implementation cost of €3.4 billion by 2030, while creating an estimated 100000 jobs. Emission reductions are also expected to lead to increased crop yields equivalent to €230 million of product.

The Eco-design Directive is expected to lead to emission reductions of 37kt of PM and 19kt of OGC per annum by 2030, significantly improving urban air quality. This directive is expected to lead to savings equivalent to 12 % of EU electricity consumption and a reduction of dependency on energy imports of 23 % and 37 % for natural gas and coal, respectively.

List of publications

1. J. Geršl, S. Knotek, Z. Belligoli, R.P. Dwight, R.A. Robinson, M.D. Coleman (2018), Flow rate measurement in stacks with cyclonic flow – Error estimations using CFD modelling. Measurement, Vol. 129, December 2018, Pg. 167-183. https://doi.org/10.1016/j.measurement.2018.06.032

2. T. O. M. Smith (2018), Modelling a quality assurance standard for emission monitoring in order to assess overall uncertainty. In: A. B. Forbes, N. F. Zhang, A. Chunovkina, S. Eichstädt, F. Pavese (eds.), Advanced Mathematical and Computational Tools in Metrology and Testing XI. Series on Advances in Mathematics for Applied Sciences, Vol. 89, Pg. 341-348. https://doi.org/10.1142/9789813274303_0034

3. Z. Qu, O. Werhahn, V. Ebert (2018), The thermal boundary layer effects on line-of-sight TDLAS gas concentration measurements. Applied Spectroscopy, Vol. 72 (6), Pg. 853-862. https://doi.org/10.7795/EMPIR.16ENV08.JA.20200130

4. A. van Hove (2018), Towards effective emission regulations: A numerical and experimental study on flow measurement uncertainty in stacks. Masters thesis. https://doi.org/10.4233/uuid:f56ff5c1-8a9c-4b5c-b404-ab421b7ede59

5. Z. Qu, O. Werhahn, V. Ebert (2019), Mid-IR ICL based TDLAS spectrometer dedicated to monitor HCl concentration in combustion emissions. Physikalisch-Technische Bundesanstalt (PTB). https://doi.org/10.7795/810.20191119A

6. Z. Qu, O. Werhahn, V. Ebert (2019), Towards a TDLAS based optical gas standard for the absolute HCl measurements in flue gases from combustion process. Physikalisch-Technische Bundesanstalt (PTB). https://doi.org/10.7795/810.20191105

7. Z. Qu, O. Werhahn, V. Ebert (2020), The spatial heterogeneity effects on dTDLAS-based CO sensor for industrial emission monitoring applications. Physikalisch-Technische Bundesanstalt (PTB). https://doi.org/10.7795/810.20200114

8. S. Knotek, M. Workamp, J. Geršl, M. D. Schakel (2020), Narrow stack emissions: Errors in flow rate measurement due to disturbances and swirl. Journal of the Air & Waste Management Association, Vol. 71 (1), Pg. 46-59. https://doi.org/10.1080/10962247.2020.1832621

9. F. D'Amato, S. Viciani, A. Montori, M. Barucci, C. Morreale, S. Bertagna, G. Migiavacca (2020), Spectroscopic Techniques versus Pitot Tube for the Measurement of Flow Velocity in Narrow Ducts. Sensors, Vol. 20 (24):7349. https://doi.org/10.3390/s20247349

10. Z. Qu, J. Nwaboh, O. Werhahn, V. Ebert (2021), Towards a dTDLAS-Based Spectrometer for Absolute HCl Measurements in Combustion Flue Gases and a Better Evaluation of Thermal Boundary Layer

Effects. Flow, Turbulence and Combustion, Vol. 106, Pg. 533-546. https://doi.org/10.1007/s10494-020-00216-z

11. Z. Qu, J. Nwaboh, O. Werhahn, V. Ebert (2020), Effects of the spatial heterogeneity of gas matrix and thermal boundary layers on absolute TDLAS HCl measurements in hot flue gases. Physikalisch-Technische Bundesanstalt (PTB). https://doi.org/10.7795/810.20200309
12. J. Meléndez, G. Guarnizo (2021), Fast Quantification of Air Pollutants by Mid-Infrared Hyperspectral Imaging and Principal Component Analysis. Sensors, Vol. 21 (6):2092. https://doi.org/10.3390/s21062092
13. Z. Qu, J. Nwaboh, G. Li, O. Werhahn, V. Ebert (2021), Measurements of N_2, CO_2, Ar, O_2 and Air Pressure Broadening Coefficients of the HCl P(5) Line in the 1–0 Band Using an Interband Cascade Laser Applied Sciences, Vol. 11 (11), 5190. https://doi.org/10.3390/app11115190
14. F. D'Amato, S. Viciani, A. Montori, A. Lapini, I. Fraboulet, J. Poulleau (2020), Optical detection of ammonia inside a stack: Comparison of different techniques. Measurement, Vol. 159. https://doi.org/10.1016/j.measurement.2020.107746

This list is also available here: https://www.euramet.org/repository/research-publications-repository-link/

Project start date and duration:		01 June 2017, 42 months
Coordinator: Rod Robinson, NPL Project website address: http://empir.npl.co.uk/impress/	Tel: +44 208 943 7146	E-mail: rod.robinson@npl.co.uk
Internal Funded Partners: 1 NPL, United Kingdom 2 CEM, Spain 3 CMI, Czech Republic 4 PTB, Germany 5 RISE, Sweden 6 VSL, Netherlands 7 VTT, Finland	External Funded Partners: 8 CNR, Italy 9 DTI, Denmark 10 DTU, Denmark 11 ENEA, Italy 12 INERIS, France 13 ISSI, Italy 14 TU Delft, Netherlands 15 UC3M, Spain	Unfunded Partners:
RMG1: IMBiH, Bosnia and Herzegovina (Employing organisation); NPL, United Kingdom (Guestworking organisation)		