Final Publishable JRP Summary for ENV60 IMPRESS
Metrology to underpin future regulation of industrial emissions

Overview

The cost of air pollution from the 10,000 largest polluting facilities in Europe is around 150 € billion per year. The European Climate Change Programme is targeting an 80-95 % reduction in greenhouse gas emissions by 2050 compared with 1990 levels, but currently Europe is not on track to meet this target. This means that more stringent industrial emission limits are needed, along with new measurements to enable the required increased monitoring and enforcement.

This project developed measurement and monitoring technologies for industrial emissions, methods and guidance to support industry, regulators and standardisation committees. Issues such as traceable stack emission measurements and the challenges of lower emission limit values were addressed. The results have helped to removed many of the current obstacles in the way of emissions reporting and to control emissions within the framework of tighter regulation.

Need for the project

Accurate measurement of emissions of pollutants and greenhouse gases in the atmosphere is vital for enabling the control and reduction of air pollution and to protect European citizens and the environment. The emission of pollutants from industrial sources has a direct impact on air quality. Therefore the EU's long-term objective as stated by the European Environment Agency is to achieve levels of air quality that do not result in unacceptable impacts on, and risks to, human health and the environment.

The Industrial Emissions Directive (IED) 2010/75 has resulted in more stringent regulations than previously existed for key pollutants. This means that there is a greater need for improved monitoring of emissions from industrial sites such as smoke stacks, landfill sites, large scale plants and oil refinery pipelines.

Different types of measurement techniques and associated methods are needed for different types of industrial site. For exmaple, point emission measurement techniques are used where the position of the emission is known, as in the case of a stack. Whereas, open path techniques are used for area sources where it is unclear where the emission is coming from, as with a landfill site. Fugitive emissions (a subset of area emissions) are leaks from pressurised equipment such as around the many flanges used in miles of gas pipework.

The EU's Industrial Emissions Directive and the associated Best Available Technique Reference Documents are introducing lower emission limit values, in some cases requiring measurements not achievable by current standard methods. The regulations require industry to measure and report emissions, such as assessing stack emissions against concentration limit values, to report annual mass emissions, and to determine emissions of greenhouse gases from area sources.

These more stringent regulations mean that improved technologies, methods and protocols are required by industry, regulatory authorities, equipment suppliers and stack monitoring providers.

Scientific and technical objectives

The project addressed the metrology needs in emissions monitoring, and supports regulation of industrial emissions, with the following objectives:

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1. Development of alternative methods / techniques to traceably calibrate (in-situ) on-line stack monitoring instrumentation
2. Characterisation of uncertainties associated with combining infrequent, independent flow measurements with continuous concentration data
3. To validate facilities that are able to test sampling proficiency
4. To develop improved and robust remote sensing techniques for fugitive emissions (i.e. emissions from pressurised equipment due to leaks or other unintended / irregular releases of gases, mainly from industrial activities).
5. To develop a suite of metrologically robust protocols / standards covering the use of open path techniques

Results & Conclusions

Objective 1 – Development of alternative methods / techniques to traceably calibrate (in-situ) on-line stack monitoring instrumentation

Investigations were carried out on two different spectroscopy techniques that have the potential for portable, routine use for the calibration of on-line stack monitoring instrumentation: (1) Fourier transform infrared (FTIR) spectroscopy and (2) tunable diode laser absorption spectroscopy (TDLAS). Associated methods were developed for both techniques, such that where a method is a written protocol it provides sufficient guidance to ensure a certain level of quality control for the FTIR or TDLAS technique.

Testing of portable FTIR was carried out at the NPL Gas Stack Simulator facility to determine if the technique could be used to provide equivalent (in accordance with the statistical tests of CEN/TS 14793) emissions monitoring data to the recognised standard reference methods for inorganic pollutants. Water vapour was also tested as reported emissions data are required to be corrected to dry conditions. Tests were carried out that generated complex gas mixtures across concentration ranges for gases CO (carbon monoxide), NO (nitric oxide), SO_2 (sulphur dioxide), HCl (hydrogen chloride), and H_2O (water vapour) applicable to a range of common industrial processes. The results demonstrated that across all concentration ranges FTIR produced emissions data that were statistically equivalent, and so could be used in place of the standard reference methods for enforcement of emission limits under the EU's Industrial Emissions Directive. The only exception was NO where above a concentration of 200 mg.m^{-3} FTIR was found not to be equivalent [1]. The results for FTIR therefore support its use for the portable, calibration of (in situ) on- line stack monitoring instrumentation.

TDLAS experiments were carried out to determine the sensitivity of this technique to variations in temperature and pressure. The results showed that the assumption that is currently made, that the temperature and pressure profile across the stack diameter can be spectroscopically accounted for by a linear summation, is incorrect. The magnitude of the effect is of the order where either manufacturers need to include a spectroscopic correction or account should be made in associated uncertainty budgets [2].

In summary, The project has validated two portable and on- line techniques (i.e. FTIR and TDLAS) and produced two associated protocol documents that can be used to ensure minimum levels of Quality Assurance/ Quality Control (QA/QC) are achieved for emissions. These techniques and associated methods can be used by ISO 17025 test laboratories (often referred to as stack testing organisations) for carrying out calibrations of in-situ instrumentation. In support of this, the Environment Agency for England have already permitted the use of portable FTIR in the emissions sector based on the FTIR equivalency test data from this project.

Objective 2 – Characterisation of uncertainties associated with combining infrequent, independant flow measurements with continuous concentration data

In-situ instrumentation that is used to continuously measure emitted concentrations of pollutants are commonly used in many process types. However, an in-situ capability that can also measure flow is lacking and therefore a flow measurement is often provided only once a year by a commissioned test laboratory. In order to meet regulations, the process plant operator is responsible for combining concentration and flow data to give annual mass emissions for reporting to national emission inventories. However, there are often errors in these data, and so guidance is needed to ensure that the combination of uncertainties from continuous concentration data with periodical flow data is handled correctly.

This objective used modelling techniques to improve the understanding of flow uncertainties, and provided guidance on the propagation of continuous and periodical uncertainties for annual mass emission measurements.

A computational fluid dynamics (CFD) model was developed to look at stack flow uncertainty. Flow measurement in a stack can be challenging as often the gas molecules / particles are not all travelling in a straight line. Swirl is a common effect; generally the waste gas from the plant is fed into the bottom of the stack at a perpendicular angle. Thus as the waste gas flows horizontally into the bottom of the stack it hits the far side of the stack and is inverted back on itself causing swirl as it ascends. The CFD model was used to simulate flow for 3 input configurations; straight pipes, 90° elbow and 90° out of plane elbow. It was found that errors in flow measurement with S-type pitot tubes (designed for the low measurement of dirty, particulate laden air or gas streams typical in a smoke stack) could reach as much as 5% in all 3 input configurations due to the amount of swirl generated.

Based on these results, an industry guidance document was written, addressing the issue of the propagation of continuous concentration and periodic flow uncertainty sources. The aim of the guide was to provide guidance for the correct approach when estimating uncertainties associated with annual mass emissions.

The objective was successfully met by the project and has resulted in an industry guidance document 'Framework for determining uncertainty sources and the propagation of uncertainty contributions in reported annualized mass emission' which can be used by test laboratories, national regulators and process plant operators and is available for download on the project website.

Objective 3 – Validated facilities for test sampling proficiency

Stack measurements are currently carried out using either portable instruments (FTIR and TDLAS from objective 1 being such examples) or by 'wet chemistry', where stack gas is pumped out of the stack and bubbled through an absorbing solution (e.g. hydrogen peroxide is used to collect SO_2) which is then later analysed (e.g. by ion chromatography) in a chemistry laboratory. Proficiency testing of analysis is relatively easily carried out and there are many schemes that provide either gas cylinders or liquid samples for end users to measure. However, such schemes do not test sampling proficiency and due to the complex chemistry of stack emissions sampling it is a significant part of the measurement problem. Hence, a capability to test sampling was needed as this is not only critical to proficiency testing but also in developing documentary standards and new instruments.

Two new national facilities were designed, constructed and validated: a Gas Stack Simulator at VSL in the Netherlands and a Particulate Simulator at NPL in the UK. The former facility is able to generate gas mixtures and is linked to VSL’s existing wind tunnel capability allowing high precision control of the flows delivered. The NPL Particulate Simulator facility is designed to deliver synthetic dust where particle size can be controlled across a concentration range of 10’s $mg.m^{-3}$ down to the sub 1 $mg.m^{-3}$ level. The facility was designed to deliver particle sizes of <10μm as such sizes are typically emitted from stacks. Furthermore, such sizes are of significance for human health as they are able to penetrate down into the alveoli within the human lung.

In addition to this, a unique European data set was created compiling large amounts of data from German, UK and Netherland’s proficiency testing schemes for the emissions monitoring community. This data set provides a unique tool encompassing a representative sample from the European emissions monitoring community and can be accessed to determine the ‘real-world’ capability of emissions monitoring. Analysis of the data found that across the key pollutants of NO_x, SO_2, CO, VOCs (volatile organic compounds) and dust there was little change in terms of performance going back to 2003, the first year of monitoring of the three national schemes [3].

In conclusion the two facilities developed by the project are already in use on new research projects. These facilities will enable future testing and development of documentary standards and will provide the capability to instrument manufacturers to test future innovations under realistic conditions. The development of proficiency testing schemes will take some time, but once on-line they will provide invaluable data to national accreditation bodies and regulators for assessing measurement capability.

Objective 4 – Improved and robust remote sensing techniques for fugitive emissions

The existing 'Refining of Mineral Oil Best Available Technique Reference Document' requires plant operators to validate and compare calculated emissions against measurements using open path optical techniques. The document gives Differential Absorption Lidar (DIAL) an optical system that provides measurements of airborne pollutants as one example. Previously, open path optical techniques had not been fully validated for this application. Therefore the aim of this objective was to improve and validate the use of DIAL, TDLAS and Infrared (IR) camera systems for fugitive emission measurements.

A key challenge for DIAL is determining background levels of gases, (e.g. the methane naturally present in ambient air), for subtraction from measurements. The laser pulse from the DIAL instrument is absorbed by both this and for example any methane in the plume of gas leaking from a section of pipe on a refinery. The greater the bias in the measurement of the background level the greater the impact of the accuracy of determining the leak flux rate. A mathematical approach was developed by the project that improves the ability to determine and subtract background levels present during measurement, thus improving the leak detection capability of DIAL [4].

In addition, a Controlled Release Facility that replicates gaseous emissions sources, was designed, constructed and validated for testing open path optical techniques. The facility is transportable, so can be deployed in the field and its four release nodes can be positioned to mimic expected release distributions, therefore creating a known diffuse emissions scene. The facility can also release typical industrial leak rates of key pollutants.

As part of objective 1, the work on TDLAS sensing was extended using the unique 600m open path test facility at PTB. The sensitivity of TDLAS to temperature and pressure variation across this path length was characterised. Using this knowledge, corrections for such temperature and pressure effects are now possible, allowing the measurement uncertainty to be properly quantified and the measurement accuracy to be improved.

A key parameter in the sensitivity of IR cameras for detecting leaks (in pipes or storage tanks) is the temperature difference between the leak and the measurement scene. In order to investigate this sensitivity a facility was built incorporating a chamber where gases could be mixed and then viewed against different measurement temperatures. Using this facility it was possible to develop a response model to determine the sensitivity under different measurement scene scenarios. This data was used to develop best practice which was included with the IR camera protocol document in objective 5.

This objective was successfully delivered and the performance of DIAL, TDLAS and IR cameras was robustly determined for fugitive emissions monitoring. Such techniques can be used by measurement service providers in order to help plant operators to comply with the requirements of the 'Refining of Mineral Oil Best Available Technique Reference Document'. Further to this, the Controlled Release Facility now provides a unique tool to facilitate development and testing of future innovations in open path optical detection techniques.

Objective 5 – A suite of metrologically robust protocols / standards covering the use of open-path techniques.

Complimenting objective 4, the aim of this objective was to provide measurement methods for the operation of DIAL, TDLAS and IR cameras, as part of written protocols. This QA/QC for the three techniques and protocols would then be disseminated to CEN for use as input to documentary standard for determining fugitive emissions using open path optical techniques.

The three protocols were written and tested in a series of field trials tailored for each technique. The IR camera was tested at a refinery in the Netherlands; the TDLAS protocol was tested at PTB's 600 m open path test facility; and the DIAL was tested at a decommissioned refinery in the south of France [5]. In the case of the latter the Controlled Release Facility was deployed allowing known amounts of VOCs to be released in a 'real' structural environment where meteorological conditions such as wind were affected by the local infrastructure. The three sets of field testing resulted in validated measurement protocols for the associated techniques.

These 3 methods can now be used by measurement services providers in order to help plant operators comply with the requirements of the 'Refining of Mineral Oil Best Available Technique Reference Document'.

Actual and potential impact

The project developed a robust metrology infrastructure to underpin the monitoring needed for enforcement of point (stack) and area (e.g. fugitive) emissions regulation. As part of this the project developed protocols used as input by CEN/TC 264 in draft standards, stack simulator facilities, and the development of DIAL, TDLAS and IR camera monitoring techniques.

Dissemination of results

Articles were published in six journals including Remote Sensing of Environment, Atmospheric Measurement Techniques, and the Journal of Air and Waste Management Association.

Presentations on the project were also given at six conferences including the American Geophysical Union meeting and the Conference on Emission Measurement and Air Protection.

A workshop on the application of differential absorption lidar (DIAL) for pollution emissions monitoring was hosted by NPL and the Chinese NIM and attended by 50 stakeholders.

An e-learning course on emission monitoring measurement entitled 'Uncertainty calculation of flue gas velocity and volume flow rate under EN ISO 16911-1' is available on the project website.

Contribution to standards

The work of this project has provided input to the following working groups, for inclusion in draft documentary standards:

- CEN/TC 264/WG 36, 'Measurement of stack gas emissions using FTIR instruments'. Input from the project: Protocol for the measurement of stack emissions by FTIR (objective 1).
- CEN /TC 264/WG 16, 'Reference measurement methods for NO_x, SO_2, O_2, CO and water vapour emissions'. Input from the project: Protocol for the measurement of stack emissions of SO_2 using optical techniques including TDLAS (objective 1).
- CEN/TC 264/WG 38 Determination of fugitive VOC emissions. Input from the project: Protocols for DIAL, TDL and IR camera.The determination of fugitive VOC emissions working group are operating under an official EC mandate formally requesting that CEN produce this documentary standard for the enforcement of fugitive related legislation (objective 5).

In the future all measurements by FTIR and for SO_2 using optical techniques carried out across the EU for regulatory compliance purposes will have to follow the methods produced by CEN/TC 264/WG 36 and WG16, using input from the project's FTIR and SO_2 protocol documents.

The work on modelling flow uncertainties and concentration measurement uncertainties has fed into the production of an industry guidance document 'Framework for determining uncertainty sources and the propagation of uncertainty contributions in reported annualized mass emission', available via the project website (objective 2).

Early impact

- A gas stack simulator is now available at VSL (objective 3).
- A particulate simulator is now available at NPL, designed to deliver synthetic dust where particle size can be controlled, and giving a measure of sampling proficiency (objective 3).
- A tool that can model the effect of temperature and pressure on remote sensing techniques is freely available for download from https://www.ptb.de/cms/en.html (objective 1).

- The Controlled Release Facility (a transportable facility based at NPL) provides a unique tool that has already been used by manufacturers to properly validate fugitive emission measurement techniques under the conditions that are actually present in real measurement situations (objective 4).

Future potential impact

The results of this project have fed into CEN standards including EN14181. Wider beneficiaries of the work include manufacturers of high-technology products such as remote sensing devices, who will benefit from the project's new measurement methods, standards and validated protocols.

List of publications

[1]. Coleman, M.D., Render, S., Dimopoulos, C., Lilley, A., Robinson, R.A., Smith, T.O.M., Camm, R., Standring, R., Testing equivalency of an alternative method based on portable FTIR to the European Standard Reference Methods for monitoring emissions to air of CO, NOx, SO2, HCl, and H2O. Journal of the Air & Waste Management Association, 65:8 (2015) 1011-1019.

[2]. Zhechao Qu, Olav Werhahn, Volker Ebert, The thermal boundary layer effects on line-of-sight TDLAS gas concentration measurements. To be submitted.

[3]. Coleman, M.D., et al., Combining UK and German Emissions Monitoring Proficiency Testing Data Based on Stack Simulator Facilities to determine if Increasingly Stringent EU Emission Limits are Enforceable, to be submitted to Accreditation and Quality Assurance.

[4]. Harris, P., Smith, N., Livina, V., Gardiner, T., Robinson, R., and Innocenti, F., Estimation of background gas concentration from differential absorption lidar measurements, Atmospheric Measurement Techniques, 9 (2016) 4879–4890.

[5]. Gardiner, T., et al., In-field validation of remote sensing emission measurements, to be submitted to Remote Sensing of Environment.

[6]. John C. Korsmana,*, Stefan T. Persijnb, Edgar M. Vuelbanb, VOC measurements by passive IR cameras: Operational boundaries and model estimations, submitted to Journal of Loss Prevention in the Process Industries.

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