Publishable Summary for 18NRM04 HEROES
Determining new uncertainty requirements for increasingly stringent legislative HCl industrial emission limits

Overview

This project validated aspects of the HCl Standard Reference Method (SRM, EN 1911), which is legislatively required for measuring industrial emissions, demonstrating that it is unable to meet uncertainty requirements brought in via Best Available Techniques (BAT) Conclusions documents (which set permit conditions for regulated industrial processes). The regulatory framework and the ability of national regulators to carry out enforcement interventions is compromised by this. HEROES has devised new uncertainty requirements for revising the SRM, based on results of the project. Beyond this HEROES has demonstrated the current performance of portable optical technologies as a potential replacement SRM in future. The regulator guidance document will help ensure that future operational measurements are made with compliant uncertainties. The results of this project have provided the evidence required to move forward with revision of the EN 1911 standard.

Need

Limiting emissions of HCl from industrial processes is critical, as it is acutely toxic and impacts on far more ecosystems than previously thought. The incorrect implementation of the Industrial Emissions Directive and the BAT Conclusions documents it adopts led to premature deaths / years of life lost in Europe of 13 000 and 125 000, respectively. Economically, if the legislation was not enforced, the European taxpayer may have incurred high costs (€7 – €28 billion p.a.).

The HCl emission limit for industrial processes regulated under the Industrial Emissions Directive used to be 10 $mg.m^{-3}$. However, BAT Conclusions documents have brought in increasingly stringent emissions limits impacting a range of industries (e.g. 2-6 $mg.m^{-3}$ for waste incineration, <1-3 $mg.m^{-3}$ for iron and steel production, 3-12 $mg.m^{-3}$ for power stations).

The legislatively required SRM for monitoring HCl emissions was described in EN 1911 and is based on extracting stack gas through deionised water in glass impingers before off-line analysis, generally by ion chromatography. It was a requirement that the emission measurement meets an uncertainty of 30 % (k = 2) of the emission limit, but this is not possible at limits below 10 $mg.m^{-3}$. The method uncertainty is subtracted from the reported emission prior to comparison to the emission limit, thus ensuring any breach is 'beyond reasonable doubt' (i.e. 95 % confidence) and that there is justification for national regulator enforcement intervention. Hence, if the method uncertainty was unclear then enforcement becomes untenable.

To address these issues three key needs were identified, from which the project's objectives were derived:

- CEN/TC 264 'Air Quality' have listed in their future priority work document the need for "assessment of current SRM to meet stricter limit values";
- CEN/TC 264 'Task Force Emissions – who provide recommendations to the European Commission – have stated that a regulatory guidance document was needed on, "…stationary source emissions providing information on the field of application of the methods (measurement range, validation range, uncertainty, etc.)";
- CEN/TC 264 have identified the need for work on "automated methods for measuring emissions".

Objectives

The overall objective of the project was to facilitate the monitoring and enforcement of emission limits at industrial processes regulated via BAT Conclusions documents. The specific objectives were:

1. Extending Stack Simulator capability in Europe to generate emissions in-line with industries regulated via BAT Conclusions (<10 $mg.m^{-3}$) adopted under the Industrial Emissions Directive (IED 2010/75/EU).

2. Characterised the performance of the HCl SRM (EN 1911) using the capability developed in objective 1 at BAT Conclusion processes. Characterised new uncertainty sources that became significant at low concentrations.

3. Produced a metrologically valid evidence base of uncertainty contributions to EN 1911 and from this developed a series of recommended uncertainty requirements for regulatory monitoring.

4. Created a scientific evidence base of the performance of a range of portable optical technologies, using the extended Stack Simulator capability developed in objective 1, with a view to the future replacement of the existing SRM. This underpins current work at CEN on CEN/TS16429 (optical techniques) and CEN/TC264/WI00264151 (Fourier transform infrared spectroscopy).

5. Produced a National Regulator Guidance Document of recommended measurement uncertainty requirements. Disseminated the document to EU DG Environment (Directorate-General for Environment), Task Force Emissions, CEN/TC 264 'Air Quality', and more broadly, to the emission community.

6. Contributed to a revision of EN 1911 by providing to CEN/TC 264 the data, methods, guidelines and recommendations, necessary for new uncertainty requirements for HCl industrial emission limits.

Progress beyond the state of the art

This project built, in part, on work carried out in 16ENV08 IMPRESS 2, which highlighted that uncertainties of 30 % are not possible for HCl at low emissions, developed a Computational Fluid Dynamics (CFD) model of swirling flow in stacks, and supported manufacturers by highlighting quantification errors in optical systems due to pressure gradients across the optical path length (a tool to calculate this effect allowing correction is available for download). HEROES has carried out work to provide practical solutions for the emissions community to facilitate monitoring and enforcement at industries regulated via BAT Conclusions.

Extension of Stack Simulator capability in Europe

This project developed and validated extended capability of European Stack Simulators for generating gas matrices composed of low level HCl concentrations, commensurate with industries regulated via BAT Conclusions documents. The validation data has been made publicly available through a data repository.

Characterisation of the performance of the HCl SRM (EN 1911)

This project characterised the uncertainty of the HCl SRM for BAT Conclusion industries. This was through work on flow calibration error, further CFD modelling to determine droplet distribution in a swirling flow and its impact when sampling isokinetically, and work on HCl uptake in stack sample collection as a function of glassware configuration and collection parameters.

Evidence base of uncertainty contributions to EN 1911

A new HCl SRM evidence base was compiled from the novel work above, Stack Simulator ILC (interlaboratory comparison) of stack testing organisations performing EN 1911 and an ILC of analytical laboratories carrying out the analytical element of EN 1911. This evidence base now underpins the uncertainty requirements for the extension of EN 1911 to industries regulated via BAT Conclusions and has been placed in a suitable repository locatable by Registry of Research Data Repositories (RE3DATA).

Evidence base of the performance of a range of portable optical technologies

A new evidence base of the performance of a range of portable optical technologies for BAT Conclusion industries has been compiled from a Stack Simulator trial and placed in a suitable repository locatable by RE3DATA. This underpins future elaboration of CEN/TS 16428 and WI 00264151 with a view to a long-term replacement for the SRM. Appropriate data from the evidence base was presented to CEN/TC 264 and CEN/TC 264/WG 3.

National Regulator Guidance Document

A new National Regulator Guidance Document of BAT Conclusions industries uncertainty requirements has been prepared and presented to different stakeholders e.g. IMPEL (European Union Network for the

Implementation and Enforcement of Environmental Law), Task Force Emissions, CEN/TC 264, and DG Environment.

Results

Extension of Stack Simulator capability in Europe

Across Europe there are a number of facilities capable of simulating an industrial stack, which provide controlled emissions allowing research into the effective measurement capability. NPL in the UK and INERIS in France host their own stack simulators. Validation was required to demonstrate that the simulators were capable of producing measurable and reliable HCl concentrations at the low levels required for this project. Through the validation tests both facilities have successfully extended their operating range for the generation of HCl down to 1mg.m^{-3} and 2.3 mg.m^{-3} for NPL and INERIS facilities, with the target under HEROES being <10mg.m^{-3}.

The INERIS validation exercise included cross contaminant species (NH_3 and SO_2) which can form salts with HCl, but the facility demonstrated homogeneous HCl concentration in the gas stream even when these were present (4% inhomogeneity uncertainty). The NPL facility was validated to a lower concentration using a dry gas stream before additional tests investigated different levels of water vapour, demonstrating homogeneous HCl concentration at H_2O volumes up to 11% (3.6% inhomogeneity uncertainty). This allowed these facilities to host interlaboratory comparisons (ILCs) at low HCl concentrations to assess the performance limitations of existing methodologies.

With both the NPL and INERIS facilities being verified for HCl at concentrations of 1 mg.m^{-3} and 2.3 mg.m^{-3} respectively, this is well below the 10 mg.m^{-3} target for the objective, so this objective has been achieved in full.

Characterisation of the performance of the HCl SRM (EN 1911)

VTT carried out a measurement campaign at a biomass plant, testing the performance of two sets of EN 1911 equipment in parallel. Field blanks before and after sampling confirmed that rinsing efficiency was good, confirming the assertion in section 5.3.3.3 of EN 1911 that glassware can be reused after rinsing. The testing also demonstrated that the two sampling lines used were consistent.

INERIS hosted an ILC at their test bench, with 9 participants measuring using two sets of EN 1911 equipment each. As each team took two samples it was possible to consider the variability of sampling, while also comparing the analytical laboratory variability. Expanded uncertainties provided by participants based on the GUM approach met the EN 1911 uncertainty, but when half confidence interval of reproducibility for the ILC was calculated, this did not meet the uncertainty limit. This demostrates that the GUM approach does not cover all uncertainty sources (e.g. human factors; risk of salt formation in presence of ammonia; etc.), so ILC is likely to be a better indicator of actual performance. INERIS also highlighted that a full uncertainty assessment should be made for the analysis, rather than just the repeatability as required in EN 1911. In France the national regulator already requires laboratories to estimate analytical uncertainty at a minimum of three concentration levels over the validated range, since analytical uncertainty will vary between the limit of quantification and the top of the range. The French standard NF X 43-551 uses a concentration threshold, so if concentration is <5 mg.m^{-3} the uncertainty criteria is <1.5 mg.m^{-3}.

The combination of testing from VTT and INERIS has demonstrated that assumptions about the EN 1911 method are correct and viable, while also identifying and characterising general performance of the method. This meets in full the requirements for this objective.

Evidence base of uncertainty contributions to EN 1911

VTT carried out parallel measurements with four different impinger tips to investigate the effect of differences on the sampling uncertainty. Repeatability and reproducibility were calculated and verified that the different impinger designs were equivalent. Further testing at different flow rates (1.5 l/minute and 8 l/minute) also validated that there was no significant difference in the results.

CMI carried out wind-tunnel tests to test the effect of using six different configurations of sampling probe and filter holder on an S-type Pitot and isokinetic sampling probe. Different layouts led to variation of 1.6% in the Pitot calibration, something that is not generally considered when looking at overall measurement uncertainty.

NPL focussed on testing the analytical uncertainty with a blind ILC, carried out by sending sodium chloride solutions with known concentrations to multiple labs. Other samples were taken from the NPL stack simulator, then split and sent to multiple laboratories. Deviation of the known concentration samples was very low, while

the stack measurements were more variable with many being well beyond the laboratory stated uncertainties. At lower measurement ranges (ELV of 3 mg.m^{-3}) many of the results were deviating by more than the 30% uncertainty limit specified in EN 1911, before even taking into account any sampling uncertainty.

Combining the findings from VTT, CMI and NPL on uncertainties for different sections of the measurement process, along with the INERIS and NPL test bench experiments, provides a more complete picture of the actual achievable uncertainty for using EN 1911 for HCl emission measurement, thus meeting the objective in full.

Evidence base of the performance of a range of portable optical technologies

NPL and VTT carried out parallel testing of optical techniques on a test bench and a real stack respectively. NPL were comparing different instruments on their stack simulator, while VTT investigated the effects of changing filter materials when sampling with FTIR instruments. CMI carried out computational fluid dynamic modelling work on particulate flow in a stack to demonstrate the impacts of inhomogeneous flow.

The VTT testing indicated that the different filter mediums did not make a significant difference to measured values. The CMI testing showed that there can still be significant inhomogeneity even when sample port location requirements are followed. The NPL testing focused on the measurement uncertainty at low concentrations, demonstrating that the range of instrumental methods tested would struggle to consistently meet the uncertainty requirements specified in EN 16429:2021 below 6 mg.m^{-3}. EN 16429 specifies a constant uncertainty of 1 mg.m^{-3} for ELVs less than 5 mg.m^{-3}, but the NPL tests suggested that the instruments tested would conservatively only achieve 1.5 mg.m^{-3}.

The combination of results from NPL, VTT and CMI forms an evidence base for the performance of portable optical technologies, demonstrating their potential to provide future reference measurements for monitoring low HCl emitting processes regulated via the BAT conclusions. This meets the objective requirements in full.

National Regulator Guidance Document

The EA have produced a national regulator guidance document, highlighting how the findings of all the project partners are informing best practice for HCl measurements. This will help ensure measurement providers will meet the uncertainty requirements.

Project partners have ensured that the findings of the HEROES project have been communicated to the wider emission monitoring community through the national regulator guidance document which has been shared within the emission community, ensuring that the work has significant impact on future implementation of HCl measurements. This meets in full the requirements for this objective.

Impact

The consortium has been active in many standardisation activities and has given presentations to different groups, as described below. The dissemination highlights are as follows:

- Two invited presentations given at Air Quality and Emissions conference (AQE2021):
  - ‘Measurement Uncertainty – Performance of methods at low concentrations’
  - ‘Do existing standard reference methods remain fit for purpose give increasingly stringent emission limits?’
- Presentation ‘Introducing the HEROES project’ given at SR 215 Air quality, Oct 2019, Helsinki, Finland.
- Presentation ‘Introducing the HEROES project’ given at French standardisation committee X43B Air quality - Stationary source emissions.
- Presentation ‘EURAMET Pre-Normative Metrology Research Activities’ given at CEN/TC 264 Annual Plenary, May 2019, Copenhagen, Denmark.
- Representations at: UK, French and Finnish national mirror groups; Source Testing Association.
- Two training courses have taken place as part of a 3-day course ‘Industrial Air Pollution Monitoring’, hosted by University of Leeds Faculty of Engineering and Physical Sciences entitled:
  - ‘Calculations of uncertainties in stack monitoring‘;
  - ‘Understanding instrument performance standards’
- A poster presentation was made at the International Metrology Congress in September 2021. This covered the objectives of the Heroes project and the ILC on the EN 1911 method.
- Presentation ‘Heroes project and it’s results’ given at SR 215 Air quality, Oct 2022, Finland

- Presentation at the FLOMEKO 2022 conference, "Modelling of uncertainties of an emission concentration measurement in stacks", November 2022.
- Four presentations at the Emission Monitoring conference (CEM 2023):
    - Evaluation of P-AMS for measuring HCl at the NPL stack simulator in order to meet the requirements of EN1649:2021 (Instrumental reference method for HCl) under increasingly stringent emission limit values
    - Appropriate measurement places and their importance in emission measurements
    - Does the performance of the HCl standardized reference method remain suitable in a context of lowering emission limit values?
    - Standardising proficiency testing for emission monitoring: EN 17656:2022
- Publication of the standard EN 17656:2022 Stationary source emissions – Requirements on proficiency testing schemes for emission measurements, October 2022.
- Baltic workshop presenting the outcomes of the Heroes project to an audience of regulators, stack testers, analyser manufacturer, plant operators and researchers, October 2022, Finland.
- A training course on "Performance of measurement methods: LoQ, measurement uncertainties" as part of a 3-day "Monitoring of air pollutant emissions" in-company training course, France
- Trade journal article: Pienten HCl-päästöpitoisuuksien mittaamisen haasteet- Heroes-projektin havaintoja, November 2022, Finland.
- End of project workshop for Heroes Stakeholders, January 2023, Paris, France.

Impact on industrial and other user communities

The project partners have presented findings from the project at workshops and/or training courses for industry. NPL made two presentations as part of a three-day training course in the UK on "Industrial Air Pollution Monitoring". VTT organised a workshop in October 2022 for representatives in the Baltic nations, including regulatory and industrial figures, entitled "Observations from the HEROES project". This covered a wide range of findings from the project including presentations given by other project partners. A trade journal article was also published to communicate the findings to relevant communities in Finland. In November 2022 INERIS included their HEROES ILC results as part of a 3 day "Monitoring of air pollutant emissions" in-company training course, which is usually run twice a year. EA periodically meet with Scottish and Welsh national regulators and have communicated findings from the project to ensure consistent implementation of the findings across the UK. The consortium also hosted an online end of project workshop for the whole user community, at which all findings of the project were presented by representatives of all the project partners.

Multiple presentations have been made at conferences focussed on emission monitoring: the Air Quality and Emissions conference (AQE) and the Conference on Emission Monitoring (CEM). These large-scale conferences have audiences of over two hundred delegates from across the industry, regulators and instrument manufacturers, communicating the findings of the HEROES project to the key stakeholders.

By communicating the findings of the project in this way, the wider community will be more aware of the issues around uncertainty for monitoring very low concentrations and, more specifically for HCl monitoring, areas of EN 1911 that are likely to be improved upon when it is revised. This will allow the industry to adopt best practice techniques validated by the HEROES project as soon as possible, making emission measurements better.

Impact on the metrology and scientific communities

By validating some aspects of the existing measurement methods (e.g. variations in impinger tips, different sampling head configurations, etc.), which were previously assumed to be equivalent, the project has quantified uncertainties and highlighted the areas that require the most and least focus as part of future revisions. In particular the parallel testing of optical P-AMS has demonstrated on-going issues with measurement uncertainty for monitoring HCl at the lowest concentrations under the EN 16429 methodology. Along with the work on characterising a more complete uncertainty calculation for EN 1911, covering both sampling and analysis in a more complete way, will guide future work to improve equipment and implementation of these methods.

The HEROES project was represented at the International Metrology Congress (CIM2021) with a presentation and a poster, highlighting the metrological need for the project and how it would benefit industrial emission measurement and reporting. Results were presented by CMI at the FLOMEKO 2022 conference which focuses on metrological issues around measurement of flow, since flow measurement is an important part of converting the concentration measurements into reportable mass emissions.

A peer-reviewed article by NPL in the journal Metrologia investigated the impact of different flow measurement reference methods on overall reported mass emission uncertainty. A further paper by CMI and NPL on modelling uncertainties of emission measurement will be submitted to the journal Measurement. EA and INERIS are drafting another paper on historical trends of AMS calibration using EN 1911 or P-AMS to be submitted to the Journal of the Air and Waste Management Association. NPL and INERIS are drafting a fourth paper on trends in the proficiency of accredited service providers performing measurements in accordance with EN 1911, which will also be submitted to the Journal of the Air and Waste Management Association. A final peer review publication on the uncertainty associated with measuring new low level HCl emissions associated with BAT conclusions emission limits will be submitted to the Journal of the Air and Waste Management Association.

Impact on relevant standards

One of the main aims of the HEROES project was supporting the revision of the existing standard reference method, EN 1911. Through the evidence base compiled by the project partners there is now a compelling case for this revision to take place, with a focus on improving the uncertainty quantification and how it is handled at very low measurement concentrations. One of the project deliverables is a draft uncertainty annex for the revised standard, setting out the required changes for meeting uncertainty requirements at lower emission limits.

When the EN 16429 standard was published in 2021 it provided a standard reference method for HCl measurement by optical P-AMS. However, no instruments had been validated against the requirements of the new standard. The HEROES project has produced some data on implementation and highlighted the areas where instruments need to be able to improve in order to meet the required uncertainty levels for the lowest upcoming emission limit values.

NPL and VTT contributed to the development of EN 17656 – Stationary source emissions – Requirements on proficiency testing schemes for emission measurements. This sets out methodologies for checking the performance of measurement laboratories. The INERIS and NPL test benches both host annual proficiency testing schemes, so having been validated for low HCl concentrations this can be added to these schemes in future to provide a better indication of true performance. The HEROES ILC activities highlighted that real sampling introduced far higher variability in results, than would be expected from the uncertainty analysis.

VTT, NPL and INERIS have also contributed to Task Force Emissions, working on a technical report (an informative (guidance)) to provide guidelines for the elaboration of standardised measurement methods. This will benefit the development of future reference method standards like the revision of EN 1911, by formally setting out recommendations for the structure and content. This will have significant benefits for all future standard development in this field.

Longer-term economic, social and environmental impacts

The work carried out in the HEROES project has demonstrated the level of achievable uncertainty with current measurement methods and highlighted areas where this can be improved. In order to enforce the lower emission limits introduced in BAT conclusions the measurement technologies and methods need to keep up. This project has contributed to this process, helping to support legislation that will protect the environment and human health from the dangerous effects of HCl emissions.

The work testing P-AMS systems will help the future development of new technologies in this field. No current technology (tunable diode laser (TDL), cavity ring down spectrometer (CRDS) or Fourier transform infrared (FTIR), etc.) has demonstrated the ability to perform at the required level to implement EN 16429 for the lowest concentrations. The continuing race to be recognised as the new SRM will drive further investment, then once the breakthrough has been made all ISO/IEC 17025 accredited stack testing organisations providing compliance measurements for HCl will have to replace their existing equipment. To give an idea of value, process plant operators currently spend $1 billion p.a. on their own analysers to monitor stacks, with Europe making up a significant portion of this. This gives an indication of market size if all accredited providers of HCl compliance measurements needed to acquire portable optical instruments.

With respect to stack testing organisations, whilst portable analysers are more expensive than glassware, they are less labour intensive to operate. During the lifetime of an analyser (~10 years) the cost of the analyser compared to the labour costs is relatively insignificant. Furthermore, coal fired powered stations are being phased out by several smaller biomass stations which is a convenient consequence of the ongoing shift in energy production. As a result, the workload for stack testing organisations is steadily increasing.

List of publications

Smith, T.O.M.; Robinson, R. A.; Coleman, M. D.; (2021), Monte-Carlo modelling to demonstrate the influence of alternative flow reference techniques on annual mass emission uncertainty. Metrologia. 10.1088/1681-7575/ac3564

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

Project start date and duration:	01 June 2019; 42 months	
Coordinator: Marc Coleman, NPL Tel: +44 (0)208 943 6828 E-mail: marc.coleman@npl.co.uk Project website address: http://empir.npl.co.uk/heroes/		
Chief Stakeholder Organisation: CEN/TC 264 'Air Quality'	Chief Stakeholder Contact: Dr Rudolf Neuroth (secretariat to CEN/TC 264 'Air Quality')	
Internal Funded Partners: 1. NPL, United Kingdom 2. CMI, Czech Republic 3. VTT, Finland	External Funded Partners: 4. EA, United Kingdom 5. INERIS, France	Unfunded Partners:
RMG: -		