Publishable Summary for 19ENV09 MetroPEMS
Improved vehicle exhaust quantification by portable emission measurement systems metrology

Overview

Nitrogen Oxides (NO_x) and fine particles emitted from cars with combustion engines are the leading causes of air pollution. In recognition of this, EC legislation was recently introduced for on-road type approval (TA) real driving emission (RDE) tests using portable emissions measurement systems (PEMS). However, metrological validation is lacking. In order to support the accuracy and comparability of vehicle emission values procedures for metrological PEMS characterisation (for NOx, particle number (PN) and exhaust gas flow) and the development of the associated infrastructure is required. This is particularly relevant for the accurate verification of vehicle emission limits in TA, and thus vital for (i) European vehicle manufacturers, (ii) the associated measurement device industries and (iii) the legislative bodies responsible for ensuring adequate air quality despite increasing traffic emissions.

Need

The European car industry currently provides jobs to more than 6 % of the employable population. It is a growing sector of European economy that produces a trade surplus of € 90.3 billion. Furthermore, it is predicted that for decades to come, vehicles powered by internal combustion engine will remain dominant over electrically driven ones, which in 2018 have only reached a 2.0 % share of the total registered vehicles across the EU. The burden of internal combustion vehicles to the environment has decreased in recent years thanks to stricter regulations and the implementation of more effective pollution control systems. However, these reductions have not been as large as anticipated due to emission standards not delivering the expected reductions under real-world driving conditions. As a result, this sector is still responsible for an important amount of NO_x and fine particles. According to the latest European Environment Agency (EEA) report, severe violations of World Health Organization (WHO) air quality guidelines (AQG) for Particulate Matter (PM_{10} and $PM_{2.5}$) were recorded in almost all EU Member States. Moreover, for NO_2, 88 % of concentrations observed at traffic stations were above AQG limit values.

In 2016, an additional TA test procedure called RDE test was introduced by the EC. This is an on-road test using PEMS and complements in-laboratory TA tests for light duty vehicles. This regulation was amended later to introduce conformity factors for NO_x and PN. These conformity factors establish “not to exceed” limits for on-road tests compared to laboratory tests. Currently, PEMS measurement uncertainties are expected to be considered in the conformity factors. However, a comprehensive and metrologically validated uncertainty estimation has not yet been documented. From 2020, new conformity factors will apply to TAs and therefore there is increasing need from end-users (e.g., car manufacturers) for the development of accurate and metrologically validated calibration standards and guidelines for vehicle emission on-road TA RDE testing.

Objectives

The overall goal of the project is to develop the necessary metrology for PEMS to support newly introduced vehicle emission legislation for on-road TA RDE testing. The specific objectives of the project are:

1. To develop traceable methods to validate and calibrate portable NO_x emissions measurement systems (PEMS), in particular for NO_2, for concentrations from below 10 µmol/mol up to at least 2500 µmol/mol. This should include the generation of a ‘state-of-the-art’ PEMS with respect to high accuracy reference gases, development of improved gas standards, calibration methods and uncertainty evaluations, as well as the validation of commercial NO_x-PEMS.
2. To evaluate the performance of commercial particle number (PN) PEMS by comparison with traceable PN facilities; to include the characterisation of i) linearity and counting efficiencies ii) particle size dependence (at least up to 10^4 particles/cm^3 and four sizes), iii) dynamic flow behaviour including the determination of aerosol sampling and handling effects.

3. To develop application-oriented calibration procedures and uncertainty budgets for PEMS exhaust flow meters (EFM) for relevant carrier gases and to investigate the effect of dynamic flow behaviour on PEMS uncertainty.
4. To quantify the correlation between: i) RDE-PEMS measurements and laboratory dynamometer test results, ii) individual PEMS "channels" for CO_2, CO, NO, NO_2, PN, exhaust mass flow and iii) validated 'golden' (reference) PEMS and commercially available PEMS.
5. To facilitate the take up of the technology and standards developed in the project by the measurement supply chain (instrument and car manufacturers, accredited calibration laboratories), standards developing organisations (e.g., CEN, ISO) and end users (automotive industry).

Progress beyond the state of the art

The current state of the art expresses the combined uncertainty of a PEMS device and the associated RDE test as 50 % for PN, and 43 % for NO_x. These values are portrayed in current legislation by conformity factors that are used to yield "not to exceed" values from the results of the in-laboratory dynamometer test over the current Worldwide Harmonised Light Vehicle Test Procedure (WLTP) test cycle for a given car. Hence, over the complete on-road, real-world in-use RDE test, the results should not be greater than those "not to exceed" values in order to achieve a "pass" declaration by the testing authority.

This project addresses the 3 key components of a PEMS system: i.e., modules for the determination of (i) NO/NO_2 concentrations and (ii) PN, as well as (iii) exhaust mass flow. By studying existing commercial PEMS devices and comparing the performance of their components with known and fully traceable laboratory-scale instruments, the project is developing uncertainty budgets for each of these three key parts. Furthermore, based on these uncertainly budgets the major contributing elements to the uncertainty of the PEMS device will be identified, and best practices will be developed to improve the underlying factors of these uncertainties.

To move beyond the current state of the art and provide end users with accurate reference materials of NO_2 that span the necessary range for PEMS calibration (1 – 2500 µmol/mol) with uncertainties of ≤ 1 % and to meet the required uncertainty of < 2 % for PEMS NO_2 measurements, this project will extend capabilities at both ends of the current range. Each end of the range representing its unique challenges. For examples at low NO_2 amount fractions (< 100 µmol/mol) nitric acid (HNO_3) is the most abundant impurity, and it is particularly problematic below 10 µmol/mol. At higher NO_2 amount fractions (> 100 µmol/mol and > 1000 µmol/mol), dinitrogen tetroxide (N_2O_4) is found in equilibrium with NO_2, which produces analysis challenges as the temperature conditions and the analyser used for certification, both affect the amount fraction of N_2O_4 and hence the analytical result. To achieve sufficiently low uncertainties required for accurate PEMS calibration the project will go beyond the current state of the art and develop an improved understanding of the formation and evolution of the major impurities (HNO_3 and N_2O_4), improved methodologies for characterising these impurities especially for low amount fractions of HNO_3 and improved quantification of the influence of these impurities.

In order to move beyond state-of-the-art in PN calibration and validation for PEMS, novel methods and materials are being implemented. Counting efficiency at small diameters (below 20 nm) requires the development of implementation of silver nanoparticles generator, whereas at large diameters (above 100 nm), modified setups are required. There is also need for improving multiple charge correction in order to reduce uncertainties in the counting efficiency for particles larger than 100 nm.

Current methods to validate PEMS EFM are based on chassis dynamometer tests and use a CVS unit to validate the total exhaust mass of a pollutant as an integral exhaust mass value over the test cycle. The PEMS EFM itself is calibrated in a SI-traceable laboratory which typically occurs at standardised and controlled conditions. However, during on-road type tests the PEMS EFM is exposed to dynamic flows, a wide range of temperatures (–7° C up to 500 °C), varying exhaust gas compositions, and a wide range of mass flows (10 - 3000 kg/h). This project will progress beyond the current state by quantifying PEMS EFM uncertainty components under real operating conditions comprising dynamic mass flow, elevated exhaust gas temperatures, and variable chemical compositions, and by providing the PEMS EFM with an uncertainty budget representative for real operating conditions of the RDE TA tests.

Based on the deeper understanding of the uncertainty sources, this project is developing a 'golden' PEMS instrument, which will represent the best available level of accuracy that can be achieved and will use the procedures developed in the project. This qualification will make use of the project's newly developed gas and particle standards, optical transfer standards, exhaust mass flow standards and metrologically sound calibration procedures. It will be based on a commercial instrument that will be validated with available procedures on a chassis dynamometer and constant volume sampler (CVS) system set-up. This will maximise transfer and applicability for end-users. Each of the main system components (NO_x, PN, EFM) for the 'golden' PEMS instrument are to be recalibrated by the partners' best capability calibration procedures on that particular module. After the completed recalibration, the instrument will next be validated against SI-traceable transfer standards and independent measurement methodologies. Including an additional dyno test, as well as on road using the project's newly developed RDE procedures.

Using single devices qualified to become a 'golden' PEMS, this project will go beyond the state of the art by designing and facilitating a 'golden' PEMS architecture that will support the dissemination of the achieved accuracy of the golden PEMS to further transfer PEMS standards and subsequent standard PEMS. In practice this architecture will consist of comparisons of commercial PEMS, set up according to the manufacturers' guides which the project will then compare against the 'golden' PEMS.

Results

Objective 1: Extending amount fraction capabilities of high accuracy primary reference materials of NO_2

The NO_2 working range covers scenarios for both exhaust emissions from normally functioning engines (1 – 10 µmol/mol) to catalyst failure (1000 – 2500 µmol/mol). To achieve these challenging uncertainties improved methods to detect, quantify and minimise the presence of the key impurities (H_2O, HNO_3) are needed. New facilities and analytical methods have been developed at LNE and are currently being developed at NPL for the measurement of low amount fractions of HNO_3. VSL have determined a method for the quantification of N_2O_4 as well as HNO_3 using FTIR spectroscopy. PTB and VSL have shared a report on methods to determine N_2O_4 and NO_2 using FTIR spectroscopy, which NPL have implemented. NPL have designed and built a new automated evacuation and filling rig to facilitate the automated flushing and filling of cylinders to remove residual water from the cylinders prior to use and this is currently in the testing phase. VSL and Air Liquide have conducted a theoretical thermodynamic and kinetic study of the reversible gas phase dissociation of N_2O_4 to NO_2, between 15 – 30 °C, and at pressures between 800 – 1200 mbar. VSL compared experimental measurements of N_2O_4 in NO_2 reference standards with the models from the study. PTB, together with VSL, also performed a study to predict the conversion from NO_2 to N_2O_4 as a function of sampling conditions, and NPL have developed a kinetic model to predict the amount fraction of N_2O_4 for difference pressures, NO_2 amount fractions and temperatures. PTB is developing a traceable calibration method for NO_x measurements by quantifying the composition of NO_2 reference standards and evaluating the N_2O_4 dimer contribution.

NO_2 primary reference materials have been prepared in an air matrix and 12-month stability studies are being carried out. Multicomponent mixtures containing NO_2, NO, CO and CO_2 have also been prepared and 12-month stability studies are nearly complete.

NPL have developed a portable dynamic dilution system based on sonic nozzles to generate dynamic reference standards of NO_2, which will be used to underpin measurements of NO_2 at low amount fractions. The characterisation of calibration biases due to matrix gas composition (N_2) compared to exhaust gas composition (CO, CO_2) is being investigated for the 'golden' PEMS using a sonic nozzle-based dilution system.

TU-DA has upgraded and modified a dTDLAS spectrometer for use with PTB's test environment. An additional port for measuring NO_2 has been added to enable measurement of NO and NO_2, and modifications have been made to enable the spectrometer to be used as a mobile transfer standard. Testing and validation has been planned by TU-DA, VTT and PTB for a laboratory campaign at PTB and an RDE measurement campaign at VTT.

Objective 2: Metrological validation and performance tests of PN-PEMS devices

A reference soot aerosol source has been characterised for particles of diameters between 20 and 200 nm. The source consists of a diffusion flame generator operated at fuel-lean conditions and a thermal/catalytic treatment, which removes the remaining organic material. Fuel-lean conditions guarantee a more reproducible particle generation, which has been confirmed by NPL and PTB counting efficiency intercomparisons.

A traceable counting efficiency calibration method was realised at PTB using monodisperse miniCAST soot and a conditioning setup with a coagulation tube. The setup and protocols were made available to the consortium. Additionally, linearity checks were realised up to concentrations of 800k particles/cm^3. This was possible using polydisperse miniCAST aerosol particles with a count mean diameter (CMD) of 50 nm. Both tests, counting efficiency and linearity, were reproduced at NPL, although counting efficiency was only possible up to 100 nm CMD at NPL. The tests are planned to be reproduced at METAS and LNE in the next measurement round.

Volatile particle removal tests were already performed in a metrological-sound way at NPL and are pending to be validated by METAS and PTB.

Regarding particle penetration, NPL was able to quantify particle concentration reduction factors (PCRF) and particle penetration efficiency (η_{pen}) for the golden PEMS PN sensor. The latter one varied between 41 and 71 %, being lower for lower CMD particles due to diffusion losses in the instrument's tubing. In order to obtain η_{pen}, it was necessary to determine the gas dilution factor. This was realised by measuring CO_2 mole fractions up- and downstream using certified CO_2 5 % in N_2.

Objective 3: Application-oriented PEMS EFM calibration procedures and uncertainty budgets

An extensive literature study was completed to summarise the current state of the art for (SI-traceable) calibration of the exhaust flow meter, which led to a comprehensive understanding of the current measurement uncertainty of EFM calibration procedures. A generic uncertainty budget comprising all identified uncertainty sources of the EFM, as identified in literature, was created. This budget is applicable to the EFM in on-road conditions. The generic uncertainty budget clearly distinguishes between uncertainty components that are quantified in literature, and uncertainty components that are identified, but lack quantification. In addition, uncertainty components that are studied in the project in order to quantify their contribution to the total EFM uncertainty are also clearly indicated. This work was made publicly available through a deliverable (report) on Zenodo.

In a first round of experiments, partners performed tests and (traceable) calibrations using the same EFM which enables to compare dynamometer validation, CVS validation, RDE-tests, and SI-traceable calibration of the EFM. The experiments led to a quantitative measure of practical measurement uncertainty of the EFM. Experiments were performed indicating potentially strong effects from vibrations created in certain conditions on the EFM error, i.e., EFM errors exceeding 10 %.

A second round of experiments is currently ongoing using the 'golden' PEMS EFM. An SI-traceable calibration was performed, as well as CVS validations in stationary and dynamic mode. Furthermore, experiments to quantify the effect from electromagnetic radiation and ambient temperature were completed. The results will be analysed to provide quantitative information on the uncertainty contributions from dynamic flow effects, temperature, and electromagnetic radiation.

Objective 4: Real-world assessment of PEMS performance

The assessment of the PEMS devices continued by performing further dynamometer/CVS and RDE measurements with the corresponding PEMS devices selected for the project. The assessment during this period included two test phases: the final round of testing of the commercial PEMS devices and the initial assessment for the golden PEMS as in factory calibrated conditions, as well as in aged condition.

The performed tests with the commercial PEMS units have been focused on studying how aging affect the determination of CO, CO_2, NOx, and PN concentration. Ageing affected the drift in CO_2 and CO determination. However, no significant ageing effect was found for NO_x and PN concentration. A comparison between commercial PEMS units resulted in PN analysers based on diffusion charging to be less sensitive at low concentrations. The measurements also resulted in satisfactory comparison for NO amount fraction, but significant differences were observed for CO and CO_2 responses. NO_2 amount fractions were challenging due to the low amount fraction measured (around 10 ppm).

Impact

Project outputs have been presented in different conferences, including Flomeko 2022 where the summary report on the current state-of-the-art of Exhaust Flow Meter (EFM) calibration procedures was introduced to the community. Additionally, the project goals and achievements have been presented at the ETH conference on combustion generated particles, a high-visibility conference for PEMS end-users and manufacturers.

The project outputs are also being disseminated to the stakeholder community via the website https://metropems.ptb.de. Additionally, dissemination is also taking place via LinkedIn.

Metadata and reports generated in the project are openly made available on zenodo, in a dedicated *community* available on https://zenodo.org/communities/metropems/.

A stakeholder workshop was held at the start of the project in which important feedback was gained from stakeholders such as the need to compare EFM experimental data with on board data acquisition data and the confirmation that uncertainty from dynamic flow effects needs to be further quantified. Two more workshops are planned for M35 and M36.

A peer-reviewed publication on the characterisation of a silver particle generator to be used for aerosol classifier calibration has been published as an open-access article in the Journal of Aerosol Science (https://doi.org/10.1016/j.jaerosci.2022.105978).

Impact on industrial and other user communities

The most recent regulations on RDE for TA have put significant pressure on research and development throughout the whole automotive manufacturing supply chain. This project is developing the necessary traceable calibration methods as well as a support infrastructure for NO_x/NO_2 (objective 1), PN emission measurements (objective 2), and EFM (objective 3). These traceable calibration methods are being tested in real-world applications (objective 4) and calibration guidelines are being produced. The project's stakeholder committee is regularly providing feedback and guidance for the project. As a result, the project's outputs are already benefitting industrial end users and products by providing enhanced quality to measure vehicle exhaust emission.

This project is also significantly extending the measurement capabilities of the participating NMIs, by the development of a support infrastructure for NO_x, PN and EFM. These capabilities will include improved flow calibration services for PEMS exhaust flow for partner VSL. New measurement services for primary reference materials of NO_2 from 1 – 2500 µmol/mol for partners NPL and VSL. New calibration services for PN-PEMS for partners PTB, NPL and METAS. In addition, PTB will introduce a 'golden' PEMS calibration service for use by end-users, which has been already calibrated using primary standards in the different NMIs.

The project's outcomes are being disseminated to stakeholders and industrial end users through public training courses and workshops focusing on good practices and methods for PEMS calibration. In addition, via publications at scientific conferences and in scientific journals as well as via trade journals.

Impact on the metrology and scientific communities

The partners in this consortium are active in the BIPM Consultative Committee for Amount of Substance: Metrology in Chemistry and Biology (CCQM) Working Group on Gas Analysis (GAWG), EURAMET Technical Committee of Metrology in Chemistry (TC- MC) and CEN TC 301/WG 6, subcommittee on Gas Analysis, therefore the outputs of this project have been presented to both groups. The improvements of high accuracy primary reference materials of NO_2 and the validation of PEMS PN measurements are already being used in activities of the metrology community to assure comparability among SI traceable standards, e.g., via key comparisons. Improved NO_2, PN and exhaust flow calibration and measurement capabilities will be made available to the scientific community via partners NPL, PTB and VSL.

The main outputs of this project are improved standards for the determination of NO/NO_2 concentrations, PN and exhaust mass flow (objectives 1-3), real-world comparison of PEMS performance (objective 4) and the production of calibration guidelines for PEMS. The improved calibration standards will include static and dynamic references, as well as transfer standards. These improved reference standards and the real-world assessment of PEMS performance will significantly advance and improve confidence in the traceability of PEMS measurements.

Impact on relevant standards

The TCs predominantly targeted by this project are ISO/TC 158 (Gas Analysis), ISO TC24/SC4, WG12 on aerosol measurements and CEN TC 301/WG16 (Road vehicles), as well as the related national standardisation committees in gas and particle analysis i.e., UNECE WP29 Global RDE IWG, DIN NA 052- 00- 34-53 AK, Performance assessment of PEMS and DIN NA 062-05-73 AA, Gas Analysis and Gas Quality. This is where the work performed in this project is the most relevant and where it will have the most impact. The partners are members of these committees and are regularly participating and ensuring that the knowledge developed within the project is fed to the committees. This interaction helps to disseminate the project's outputs directly into the standardisation activities, and the requirements emerging from the standardisation committees will be used to refine the project in order to maximise its impact.

Longer-term economic, social and environmental impacts

Vehicle emissions contribute to atmospheric PM, NO_x and tropospheric ozone pollution, which in turn affect the climate, human health and agricultural yields. In particular, diesel combustion vehicles produce a significant amount of NO_x. Stricter European regulations have been established to tackle emissions, but current real-driving emission measurements for type-approval of light-duty vehicles introduce different metrological challenges. By providing high-quality reference standards and traceability to PEMS measurements, this project will support the improvement of air quality across the EU and will potentially facilitate their uptake by other countries that adopt similar TA tests for light-duty vehicles.

In addition to the environmental benefits, economic and social impacts are also expected. The European car industry is a growing sector that provided the EU with a trade surplus of € 90.3 billion in 2018. The same year, more than 15 million light-duty vehicles were registered in the EU, of which 56.7 % are fuelled with gasoline and 35.9 % with diesel. The European TA regulations require these vehicles to be tested by RDE PEMS for TA. Therefore, the creation of calibration guidelines and a metrological infrastructure for such testing will support its role out and provide confidence in the results.

List of publications

- *Characterising the silver particle generator; a pathway towards standardising silver aerosol generation.* Irwin M., Hammer T., Swanson J., Berger V., Sonkamble U., Boies A., Schulz H., Vasilatou K., Journal of Aerosol Science 163 (2022), 105978. DOI: 10.1016/j.jaerosci.2022.105978

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

<table>
<tr><td colspan="2">Project start date and duration:</td><td>01 September 2020, 36 months</td></tr>
<tr><td colspan="3">Coordinator: Jorge Saturno, PTB Tel: +49 531 592 3217 E-mail: jorge.saturno@ptb.de
Project website address: https://metropems.ptb.de</td></tr>
<tr><td>Internal Funded Partners:
1. PTB, Germany
2. LNE, France
3. METAS, Switzerland
4. NPL, United Kingdom
5. VSL, Netherlands
6. VTT, Finland</td><td>External Funded Partners:
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8. DTI, Denmark
9. KIT, Germany
10. TU-DA, Germany</td><td>Unfunded Partners:
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