



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 NO_x, 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 (PM₁₀ and PM_{2.5}) were recorded in almost all EU Member States. Moreover, for NO₂, 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 NO₂, 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 10⁴ particles/cm³ 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 CO₂, CO, NO, NO₂, 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/NO₂ 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 uncertainty 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 NO₂ 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 NO₂ 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 NO₂ amount fractions (< 100 μmol/mol) nitric acid (HNO₃) is the most abundant impurity, and it is particularly problematic below 10 μmol/mol. At higher NO₂ amount fractions (> 100 μmol/mol and > 1000 μmol/mol), dinitrogen tetroxide (N₂O₄) is found in equilibrium with NO₂, which produces analysis challenges as the temperature conditions and the analyser used for certification, both affect the amount fraction of N₂O₄ 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 (HNO₃ and N₂O₄), improved methodologies for characterising these impurities especially for low amount fractions of HNO₃ and improved quantification of the influence of these impurities.

In order to move beyond the 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 implementation of a silver nanoparticle 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 will develop and intends to qualify 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 also 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 be done in order to maximise transfer and applicability for end-users. Then, each of the main system components (NO_x, PN, EFM) for the 'golden' PEMS instrument will 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 NO₂

The NO₂ 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 (H₂O, HNO₃) are needed. New facilities and analytical methods have been developed at NPL and LNE for the measurement of low amount fractions of HNO₃. VSL have determined a method for the quantification of N₂O₄ as well as HNO₃ using FTIR spectroscopy. PTB and VSL have shared a report on methods to determine N₂O₄ and NO₂ 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. NPL have also developed a kinetic model to predict the amount fraction of N₂O₄ for difference pressures, NO₂ amount fractions and temperatures. Discussions have taken place to agree on the matrix of the NO₂ reference materials and to establish a schedule for the mixtures to ensure that if required by other partners or in other work packages they will be available. Air Liquide have prepared mixtures of NO₂ at 1, 10 and 2000 μmol mol⁻¹ and these have been shipped to NPL to conduct the stability analysis.

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.

A novel multiple charge correction (MCC) method is being developed and implemented in order to improve the counting efficiency tests required by PN-PEMS.

A Silver Particle Generator (SPG) has been tested and characterised by METAS. The generator is able to produce particles in two different size modes: below 10 nm, and above 10 nm. These particles will be used to calibrate Differential Mobility Analysers (DMAs) below 20 nm, which are the major source of uncertainty in setups for calibration of counting efficiency.

The volatile particle removal efficiency of PEMS units will be tested with tetracontane particles with geometric mean mobility diameters of 30 nm as test aerosols for particle number concentrations ≥10,000 cm⁻³.

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 will lead 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 was created.

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. It further leads to a quantitative measure of practical measurement uncertainty of the EFM and possibilities to quantify uncertainty from dynamic flow effects. Experiments were performed indicating potentially strong effects from vibrations on the EFM error, i.e., EFM errors exceeding 10 %.

Objective 4: Real-world assessment of PEMS performance

By M18, an initial assessment of one of the commercial PEMS-devices (the only unit at that time available) was performed. The assessment included tests in lab (dyno/CVS) together with RDE testing. These results represent the initial reference for one of the commercial PEMS units before service (aged). During autumn 2021, a second set of dynamometer/CVS tests was repeated for two commercial PEMS units after their annual service. After these tests, one of the commercial PEMS units was sent to another project partner for further RDE testing (and to be compared with a third commercial PEMS unit). During spring 2022, retests with the commercial units in *aged* state will be performed and the change in uncertainty will be analysed.

The performed tests with the commercial PEMS units may be concluded as follows:

Aged vs freshly calibrated PEMS tests results:

- CO₂ and NO_x values remained consistent and within the permissible tolerances throughout all tests.
- CO tolerances indicate improvements over service due to improvements in accuracy for lower concentrations.
- PN results were found consistent for higher concentrations, meanwhile the tolerance increased after service for lower concentrations. However, for the tests resulting in low concentration, the relative difference between CVS and the PEMS unit was in general found in the order of $1 \cdot 10^9$ particles/cm³ (permissible tolerance $1 \cdot 10^{11}$ particles/cm³).

During the comparison of the two commercial, freshly serviced PEMS units, the following results were concluded:

- The EFM results were found deviant for one of the PEMS devices, resulting in higher exhaust mass results for all components.
 - The temperature correction was found as the main source for the deviation.
 - Static EFM tests support these findings, as these results were compared to calculated exhaust mass flow acquired from CVS.
- Due to the difference in EFM results, the PEMS units were compared based on exhaust concentrations instead. Results for the gaseous components were statistically in agreement. PN results correlated better for the commercial PEMS equipped with a CPC compared to the individual utilising a diffusion charger. Especially low PN concentrations were found challenging for the unit equipped with a diffusion charger.

Impact

By continuous interaction with the stakeholder community and from the output of the first stakeholder workshop, the consortium has gained relevant feedback regarding, for example, the use of on board data acquisition in RDE and the implementation of novel techniques for particle generation below 23 nm, among other important topics.

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 will create impact by supporting this research and developments and by providing traceable calibration methods as well as a support infrastructure for NO_x/NO₂ (objective 1), PN emission measurements (objective 2), and EFM (objective 3). These traceable calibration methods will be tested in real-world applications (objective 4) and calibration guidelines will be produced. The consortium has good connections with PEMS manufacturers and intends to include them as part of the project's stakeholder committee (to provide feedback and guidance for the project). As a result, the project's outputs will be aimed at and will benefit industrial end users and products by providing enhanced quality to measure vehicle exhaust emission.

This project will also significantly extend the measurement capabilities of the participating NMIs, by the development of a support infrastructure for NO_x, PN and EFM. This will include improved flow calibration services for PEMS exhaust flow for partner VSL. New measurement services for primary reference materials of NO₂ 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.

The project's outcomes will be 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) and EURAMET Technical Committee of Metrology in Chemistry (TC- MC), subcommittee on Gas Analysis, therefore the outputs of this project will be presented to both groups. The improvements of high accuracy primary reference materials of NO₂ and the validation of PEMS PN measurements will support future activities of the metrology community to assure comparability among SI traceable standards, e.g., via key comparisons. Improved NO₂, 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/NO₂ 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 will be the most relevant and where it will have the most impact. The partners are members of these committees and will ensure that the knowledge developed within the project is fed to the committees. This will ensure that the project's outputs feed 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 vehicles are emitting under real-world driving conditions compared to laboratory tests and therefore current results are not fully comparable. 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. Last 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.



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| Project start date and duration: | | 01 September 2020, 36 months | |
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| Internal Funded Partners: | External Funded Partners: | | Unfunded Partners: |
| 1. PTB, Germany 2. LNE, France 3. METAS, Switzerland 4. NPL, United Kingdom 5. VSL, Netherlands 6. VTT, Finland | 7. Air Liquide, France 8. DTI, Denmark 9. KIT, Germany 10. TU-DA, Germany | | - |
| RMG: - | | | |