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 introduced for on-road type approval (TA) real driving emission (RDE) tests using portable emissions measurement systems (PEMS). However, metrological validation has not been established. This project has provided insights and developed reference materials 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). 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.

There are still ongoing challenges in providing stable reference materials for low concentration fractions below 10 µmol mol^{-1}. An improved infrastructure for in-field performance validation of RDE PEMS devices was developed using an optical gas standard device. An instrument intercomparison across three laboratories revealed discrepancies in PN measurements, particularly for particles below 30 nm, and gas matrix influenced the gas dilution factor by up to 17 %. Regarding exhaust flow, meeting the 3 % accuracy legal limit in SI-traceable calibration for two EFM units was proven feasible, experiments with pulsations indicated limitations for flow rates exceeding approximately 50 % of the maximum, although this pulsation type is uncommon in on-road conditions. The project successfully assessed PEMS performance in real-world conditions. As part of the recommendations, special attention should be given to tuning PEMS devices to specific emission matrices and to refining analyser drift criteria in current RDE regulations.

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 remains 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. From 2020, new conformity factors 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. Conformity factors are expected to be adjusted down to 1.0 with the introduction of the Euro 7 regulations.

Objectives

The overall goal of the project was 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 were:

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 34 % for PN, and 10 % 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 addressed 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 has developed 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 were identified, and best practices were 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 extended 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 went beyond the current state of the art and developed 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 were implemented. A modified calibration setup was required for determining the counting efficiency for particles of 200 nm mobility diameter. Additionally, to assess the particle penetration efficiency, a special setup was developed, and the gas dilution factor was determined using CO_2 at concentrations representative of exhaust gas.

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 provided research beyond the current state-of-the-art 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 developed a ‘golden’ PEMS instrument, which represents the best available level of accuracy that can be achieved. This qualification uses the project’s newly developed gas and particle standards, exhaust mass flow standards and metrologically sound calibration procedures. It is based on a commercial instrument that was validated with available procedures on a chassis dynamometer and constant volume sampler (CVS) system set-up. This maximises transfer and applicability for end-users. Each of the main system components (NO_x, PN, EFM) for the ‘golden’ PEMS instrument were recalibrated by the partners’ best capability calibration procedures on that particular module. After the completed recalibration, the instrument was 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.

Results

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

This objective was successfully achieved. The improvements in accuracy of the reference standards and the adoption of multicomponent calibration standards were needed to facilitate the reduction of the measurement uncertainties associated with PEMS NO_2 measurements to below 2 %, and the analytical and mobile transfer standard was required to preserve these uncertainties achieved in the laboratory to the application of the PEMS device in RDE testing. Together, these approaches provide metrological validation for PEMS measurements through more accurate calibrations and reduce the uncertainty of PEMS NO_2 measurements to below the current state of the art of 43 %. A validation report on the low amount fraction (1 – 10 µmol mol^{-1}) NO_2 primary reference standards developed has been produced, and a best practice guide on the use of high amount fraction (1000 – 2500 µmol mol^{-1}) NO_2 reference standards has been produced to share the findings with the broader NMI community and specialty gas industry.

While improvements were made to the preparation procedures to remove as much water as possible, its persistence led to NO_2 losses of 4 – 13 % over 12 months, with the major impurity confirmed to be HNO_3. As a result of the problems with the stability of low amount fraction NO_2 standards, the non-linear decay profiles, and the challenge in removing and avoiding reintroducing water during preparation and handling, an alternative approach would be to wait for most of the reaction with water to occur (100 days after preparation) and then analytically determine the amount fraction of NO_2 using a dynamic standard with a low uncertainty. In this case, it should be possible to achieve the uncertainty requirement of 2 % for NO_2.

The effect of the matrix gas composition on the analytical measurement bias for PEMS measurements was found to be very significant for both NO and NO_2 (20 % and 30 % variation in sensitivity respectively) for the PEMS device tested. During calibration with reference standards in nitrogen the response of the PEMS device was found to exhibit a non-linear relationship to both NO and NO_2. Due to this significant non-linearity, higher order calibration functions are required for accurate NO and NO_2 determination. When calibrating with NO_2 in synthetic air standards, the PEMS response exhibited a linear relationship. This change in behaviour between nitrogen and air matrix compositions poses a significant challenge in calibrating PEMS accurately and demonstrates the need for calibration gases in a matrix that closely resembles actual tailpipe emissions.

An instrument based on direct Tuneable Diode Laser Absorption Spectroscopy (dTDLAS) was developed to be used as an analytical transfer standard for NO_x measurements and demonstrated good linear agreement with NO and NO_2 reference standards during field tests, obtaining uncertainties of 10 % and 6.3 % respectively. The instrument was further upgraded and optimised for mobile application for use as a mobile transfer standard for PEMS measurements, and it was compared to the “golden PEMS” during RDE tests.

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

This objective was successfully achieved. The first NMI-level comparison of PEMS-PN counters was realised in the project and involved three different laboratories that independently measured linearity response of a CPC-based PEMS, counting efficiency, volatile particle removal efficiency, particle penetration efficiency and particle concentration reduction factor. For this comparison, a harmonised calibration strategy was developed in the project and implemented in the different laboratories. The harmonised calibration strategy was distributed to the stakeholder community and feedback from the community was considered in its preparation.

The linearity measurements for a CPC-based PEMS were realised up to a concentration of 2×10^6 cm^{-3} for poly-disperse particles of 70 nm (count mean mobility diameter, CMMD) and the slope was within the regulatory limits for all the determinations in the different laboratories. A traceability path using monodisperse 70 nm CMMD particles was proposed.

Counting efficiency was done for monodisperse particles with six different CMMD: 23, 30, 50, 70, 100 and 200 nm. The smaller the particle size, the larger the discrepancies found among the different measurements done at the different NMIs. For example, for 23 nm CMMD, a difference larger than 15 % was found, this difference was < 6% for 30 nm particles, which is within the expected range observed in previous comparisons. For particles larger than 50 nm, the three measurement results approached each other within 5 %.

The particle concentration reduction factor (PCRF) was found to be over 100 for particles of 23 nm CMMD. However, for particles larger than 50 nm CMMD, it decreased significantly, down to 85. This result implies that a constant PCRF applied to PN concentrations measured by PEMS is not an accurate approach but rather a worst-case scenario, which could lead to an overestimation of particle counts by PEMS, depending on the size distribution of the aerosol particles emitted by the vehicle.

Not included in PEMS regulation, the particle penetration efficiency (PPE) provides a more realistic approach to understand how particles are treated and determined by a PEMS system. During the project, the PPE was determined by two different laboratories by first determining the PCRF and the gas dilution factor of the system. For that purpose, certified CO_2 (5 %) was used. The resulting PPEs were significantly different, being 64 ± 5 and 78 ± 6 for 100 nm CMMD particles. These discrepancies originated from different gas dilution factors rather from PCRF measurements, which in fact were in good agreement. The gas dilution factors measured in different matrices, like CO_2 in N_2 or CO_2 in synthetic air, were significantly different according to measurements in one of the laboratories.

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

This objective was successfully achieved. Best capabilities were combined by the collaborating participants to yield the most EFM uncertainty information for on-road conditions possible: KIT investigated EFM uncertainty from dynamic flow (load) changes, gas temperature, and variable gas composition. VTT investigated the relation between CVS-validations of the EFM and the (independent) SI-traceable calibration result, and EFM uncertainty from dynamic flow (load) changes using a fast-responding fuel intake sensor and the link to the requirement set out in the RDE regulation. VSL performed the SI-traceable (check) calibrations, and evaluated the EFM uncertainties from electromagnetic radiation effects, pulsating flows, and ambient temperature effects. The information gained was used to draw conclusions on factors potentially affecting EFM measurement uncertainty in on-road conditions, to draft recommendations for improving the EFM uncertainty assessment and for proposing EFM verification improvements when using dynamometer setups.

The application-oriented calibration procedure and uncertainty budget were provided by considering that the generic uncertainty budget, including the SI-traceable EFM calibration, should be tailored to the specific EFM size, model, manufacturer, and serial number, and the circumstances in which it was operated (e.g., cold start, gasoline/diesel, dynamic flow effects, and pressure/temperature effects). The effects from dynamic flow changes and from variable carrier gases were investigated by comparison of integrated mass flows for a variety of PEMS-systems, vehicle types (petrol, diesel, and hybrid), and exhaust gas mixtures. No definite quantitative figures on uncertainty influences could be provided; rather improvements in the EFM verification procedures were proposed.

Objective 4: Real-world assessment of PEMS performance

The main motivation for the work related to real-world assessment of PEMS performance was to validate and to define factors affecting the performance of PEMS devices. Another key motivation was to study the correlation for PEMS response between laboratory and RDE measurements. This evaluation was performed both on a system level and for the individual PEMS channels, CO_2, CO, NO, NO_2, PN, exhaust mass flow. The collective goal was 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.

This objective was successfully achieved. The primary goal was to validate and identify factors influencing the performance of PEMS devices, with a particular emphasis on studying the correlation between laboratory and

Real Driving Emissions (RDE) measurements. Three main tasks were executed: identifying the state of the art for PEMS measurement uncertainty for RDE, assessing and comparing the performance of different commercial PEMS systems in both laboratory and on-road conditions, and setting up, calibrating, and evaluating the performance of a 'goldenPEMS' instrument. The findings indicate that one commercial PEMS device consistently met and exceeded RDE regulation criteria over a three-year testing period. The evaluation of PEMS performance in various test conditions and exhaust matrices revealed insights into the measurement challenges and strengths of different emission types. The correlation between laboratory validations and RDE testing highlighted variations dependent on emission and vehicle types, emphasising the need for tuning PEMS devices for different emission matrices. The results also suggested opportunities for further refinement of the analyser drift criteria defined in the current RDE regulation. Overall, the successful achievement of this objective contributes with valuable insights into PEMS performance and its application in real-world scenarios.

The performance of the tested PEMS devices varied somewhat between laboratory and RDE testing. The deviations between the two tested PEMS devices were found dependent both on emission type and emission characteristics but also vehicle type. For example, despite the CO_2 results agreed relatively well between the two PEMS types both in laboratory and in RDE, the no clear correlation for the deviations between laboratory tests and RDE could be withdrawn. Because NO_x emissions recorded from most of the vehicles were low, no systematic trends or correlations between variability for the PEMS devices were found. The performance of the tested PN PEMS analysers varied depending on vehicle type. The results recorded with ADC PN analyser correlated better with the laboratory result for tests producing lower PN emissions, meanwhile the CPC based PN device performed better for tests which resulted in higher PN emissions. The results suggest that the two PN PEMS devices may have to be tuned for different emission matrixes. The data produced both in laboratory and in RDE conditions indicated that similar trends and correlation for the recorded deviation occurred.

Impact

The project's outcomes were disseminated to stakeholders and industrial end users through public training courses and workshops focusing on good practices and methods for PEMS calibration. Three different training activities were organised during the course of the project: MetroPEMS stakeholder workshop (December 2020), MetroPEMS technical training course (August 2023) and MetroPEMS final workshop (August 2023).

In addition, the project was disseminated via publications at scientific conferences and in scientific journals as well as via trade journals such as AWE International.

Project outputs have been also presented at international conferences such as ETH conference on nanoparticles 2021, 2022, and 2023, Zurich, Switzerland; European aerosol conference 2023, Malaga, Spain; Flomeko conference on Flow measurement 2022. China; International Aerosol Conference 2022, Greece and Metrology for Climate Action conference (WMO-BIPM) 2022, online.

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 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. The project outcomes were presented to two different standardisation committees including the Particle Measurement Programme of the United Nations Economic Commission for Europe (2022 and 2023) and the EURAMET TC-Flow (2021).

The project outputs are available via the website https://metropems.ptb.de. Additionally, dissemination also took place via LinkedIn. Metadata and reports generated in the project are openly made available on Zenodo https://zenodo.org/communities/metropems/.

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 has created impact by supporting this research and developments and by providing 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 were tested in real-world applications (objective 4) and calibration guidelines were produced. The consortium has good connections with PEMS manufacturers and included them as part of the project's stakeholder committee. The project's outputs are useful to industrial end users and provide enhanced quality procedures to measure vehicle exhaust emission using PEMS.

This project extended the measurement capabilities of the participating NMIs, by the development of a support infrastructure for NO_x, PN and EFM. This includes improved flow calibration services for PEMS exhaust flow for VSL, new measurement services for primary reference materials of NO_2 from 1 – 2500 µmol/mol for NPL and VSL, and new calibration services for PN-PEMS for PTB, NPL and METAS. In addition, PTB has introduced a 'golden' PEMS calibration service for use by end-users.

Impact on the metrology and scientific communities

The partners in this consortium were 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), therefore the outputs of this project were presented to both groups. The improvements of high accuracy primary reference materials of NO_2 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 NO_2, PN and exhaust flow calibration and measurement capabilities have been made available to the scientific community via NPL, PTB and VSL.

Impact on relevant standards

The TCs predominantly targeted by this project were ISO/TC 158 (Gas Analysis), ISO TC24/SC4, WG12 on aerosol measurements and CEN TC 264 Air quality. The partners are members of these committees and ensured that the knowledge developed within the project was fed to the committees. This interaction ensured that the project's outputs were fed directly into the standardisation activities, and the requirements emerging from the standardisation committees were 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, however, it has been found that vehicles were emitting under real-world driving conditions as opposed to laboratory tests and therefore current results are not fully comparable. By providing high quality reference standards and traceability to PEMS measurements, this project supports 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 European TA regulations for light duty vehicles require these to be tested by RDE PEMS. Therefore, the creation of calibration guidelines and a metrological infrastructure for such testing is essential to provide confidence in the results.

List of publications

Hammer, T., Irwin, M., Swanson, J., Berger, V., Sonkamble, U., Boies, A., Schulz, H., & Vasilatou, K. (2022). Characterising the silver particle generator; a pathway towards standardising silver aerosol generation. In Journal of Aerosol Science (Vol. 163, p. 105978). Elsevier BV. https://doi.org/10.1016/j.jaerosci.2022.105978

Vasilatou, K., Wälchli, C., Auderset, K., Burtscher, H., Hammer, T., Giechaskiel, B., & Melas, A. (2023). Effects of the test aerosol on the performance of periodic technical inspection particle counters. In Journal of Aerosol Science (Vol. 172, p. 106182). Elsevier BV. https://doi.org/10.1016/j.jaerosci.2023.106182

Schakel, M., Stiphout, W., Pettinen, R., Yu, Z., & Wagner, U. (2023). Traceable Uncertainty of Exhaust Flow Meters Embedded in Portable Emission Measurement Systems. Elsevier BV. https://doi.org/10.2139/ssrn.4605945

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

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