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Measuring Sub-23 Nanometer Real Driving Particle Number Emissions Using the Portable DownToTen Sampling System --Manuscript Draft--

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TITLE:

Measuring Sub-23 Nanometer Real Driving Particle Number Emissions Using the Portable DownToTen Sampling System

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KEYWORDS:

automotive, emissions, particle number, sub-23 nm, portable emission measurement, real driving emissions, sampling, dilution

SUMMARY:

Presented here is the DownToTen (DTT) portable emission measurement system to assess real driving automotive emissions of sub-23 nm particles.

ABSTRACT:

The current particle size threshold of the European Particle Number (PN) emission standards is 23 nm. This threshold could change because future combustion engine vehicle technology may emit large amounts of sub-23 nm particles. The Horizon 2020 funded project DownToTen (DTT) developed a sampling and measurement method to characterize particle emissions in this currently unregulated size range. A PN measurement system was developed based on an extensive review of the literature and laboratory experiments testing a variety of PN measurement and sampling approaches. The measurement system developed is characterized by high particle penetration and versatility, which enables the assessment of primary particles, delayed primary particles, and secondary aerosols, starting from a few nanometers in diameter. This paper provides instruction on how to install and operate this Portable Emission

Measurement System (PEMS) for Real Drive Emissions (RDE) measurements and assess particle number emissions below the current legislative limit of 23 nm.

INTRODUCTION:

The Particle Measurement Programme (PMP) was founded by the UK Government for the “development of type approval test protocols for assessing vehicles fitted with advanced particulate reduction technology that would complement or replace current legislative measurement procedures”¹. The PMP is the world’s first particle number-based emissions regulation, targeted specifically at carbonaceous particles ≥ 23 nm. Recent measurements indicate that it may be necessary to include smaller particles.

Negative health impacts of diesel soot are well understood², and therefore, the ‘precautionary principle’ was invoked on the basis that the elimination of carbon particles from diesel exhaust, via the mandatory use of diesel particulate filters (DPFs), was imperative on health grounds. However, because in European legislation a limit value must force adoption of emissions control technologies, this could not be achieved without an appropriate measurement method. With strong political backing across Europe, the UK Government led the conception of the PMP to improve particulate measurements. The PMP, under the auspices of the United Nations Economic Commission for Europe (UN-ECE)³, included the expertise of others from around the globe. Two particle research projects were completed in 2001. One of them (Particulate Research⁴) was carried out by the UK Government Department of the Environment, Transport and the Regions (DETR), in partnership with the Society of Motor Manufacturers and Traders (SMMT) and the Oil Companies European Organisation for Environment, Health and Safety (CONCAWE). The other one (PARTICULATES⁵) was funded by the European Union’s 5th Framework and was carried out by 14 different European partners. The results of both projects indicated that particle number-based procedures were promising, but that challenges for repeatable and reproducible measurements remained.

In 2007 the final report of the PMP Light-duty Inter-laboratory Correlation Exercise was published⁶, including some improvements on the filter-based mass measurement method, primarily demonstrating the feasibility of a number count-based method for regulatory purposes based upon a defined particle size range and particle volatility. Both methods were implemented based upon sampling from the existing constant volume sampler (CVS) dilution tunnel approach originally developed for particulate matter mass and bagged dilute gaseous emissions measurements.

Within the number count-based method, a lower particle size limit of ~ 20 nm was selected. The primary objective of the project was to ensure particles of this size and above were controlled by legislation. It is now known that the primary particle size in engine exhaust can be < 20 nm⁷⁻⁹. For practical reasons, a particle counter with a 50% counting efficiency (d_{50}) at 23 nm was selected, and this size became the accepted lower size threshold. It was recognized that due to the high sensitivity to properties such as dilution, air temperature, humidity, and ratio¹⁰, volatile particle size distribution and integrated number measurements could be repeatable in one CVS-equipped facility with one vehicle, but much less so from facility to facility. Thus, for rigorous regulations,

it was necessary to focus purely on nonvolatile particles, with the measurement approach effectively defining the regulatory particle boundary conditions on size and volatility. European diesel fuel has back-end volatility such that only a few percent boils at temperatures above 350 °C, and early work within the PMP indicated that short residence times at this temperature were suitable for the complete evaporation of tetracontane, a linear hydrocarbon containing 40 carbon atoms with volatility towards the end boiling point of engine lubricant¹¹. Consequently, a temperature of 350 °C has become the de facto reference point for regulatory >23 nm particle volatility.

The PMP measurement system specification comprises components for sampling, sample conditioning, and measurement, summarized in **Table 1**.

[Place **Table 1** here]

The European PMP PN approach is being implemented and now applies to light-duty diesel (September 2011, EURO 5b) and GDI vehicles (September 2014, EURO 6), and to diesel and gas heavy-duty engines (February 2013, EURO VI).

Recent measurements showed that some light-duty vehicles and, in particular, spark ignition technologies, can emit substantial levels of particles <23 nm^{12,13,14}. This led the European Commission to fund research projects to develop new or extended methods that can be rapidly implemented as a replacement, or addition to, the current >23 nm regulation.

One such project, DownToTen (DTT), aims to preserve the general approach of PMP and extend the measurement range down to a $d_{50} \leq 10$ nm. To this end, the configuration of the DTT measurement system was designed to include the same basic elements described in **Table 1**, but with the conditioning and measurement steps optimized to enable efficient transport and detection of the <23 nm particles. The DTT system was initially developed for laboratory use but was modified to operate as a portable emissions measurement system (PEMS). For the DTT PN-PEMS system, the components were optimized to reduce weight and power consumption and increase physical robustness without substantially diverging from the original design. For mobile application, the system must be resistant to harsher and erratic temperatures, pressures, and vibration environments likely encountered in light- and heavy-duty PEMS testing. The impact of pressure variations at the inlet of the system was modelled and studied experimentally¹⁵. The resistance to vibrations was assessed using a dedicated test bed¹⁶. Vibrations and accelerations that occur during typical RDE drives did not impair the measurement results of the condensation particle counters used. The DTT system is also designed for use at low temperatures, where the volatile removal function is inactive, to feed an aging chamber and study secondary organic aerosol formation¹⁷.

The thermal conditioning elements of the DTT measurement system that define the regulatory volatility boundary of particles closely parallel the elements of the PMP system in that both systems contain the sequence:

1. First particle number dilution stage
2. HC/volatile elimination stage
3. Second particle number dilution stage

The primary differences between the DTT and PMP systems are that the DTT system components are selected to:

1. Maximize transmission of ~ 10 nm PN from the sample source to the particle counter using low loss dilution and particle transmission approaches
2. Comprehensively remove volatiles using oxidative particle elimination rather than merely reducing partial pressures of condensable HC species through evaporation and dilution
3. Count particles of ~ 10 – 50 nm with greater efficiency than current PMP systems

The objective of this paper is to present the use of the DTT PN-PEMS system for measuring nonvolatile particles ≥ 10 nm from an in-use road vehicle. This includes an introduction to the measurement system and its main components, performing laboratory-based calibration measurements, installing the device for a mobile application, conducting a real driving emission measurement, and processing the collected measurement data.

Instrumentation

The DTT PN-PEMS was designed to provide high particle penetration down to a few nanometers, robust particle number dilution, removal of volatile particles, and prevention of artificial particle formation. The components of the system were selected based on results from laboratory experiments comparing a variety of technologies for dilution and aerosol conditioning. This section provides an overview of the system, its working principle, and the components used. **Figure 1** shows a schematic of the system. **Figure 2** shows a photo of the system. The DTT system is 60 cm high and has a footprint of 50 cm x 50 cm. The weight of the system is approximately 20 kg. Including the required peripheral elements (i.e., battery and gas bottle) the total weight is approximately 80 kg. The major elements of system are the two dilution stages (i.e., first hot, second cold), a catalytic stripper, and at least one condensation particle counter (CPC).

[Place **Figure 1** here]

[Place **Figure 2** here]

Two dilution stages reduce the particle number concentrations to levels measurable by condensation particle counters ($< 10^4$ #/cm³). Custom-made porous tube diluters are used for both dilution stages. This technology was selected because of its low particle loss^{18,19}. The radial ingress of dilution air convectively keeps particles away from the walls, which reduces particle losses. Furthermore, these diluters can be very small and can withstand temperatures of 400 °C. The porous material used is a sintered hastalloy X tube (GKN Filters Metals GmbH,

Radevormwald, Germany). Static mixing elements inside the porous tube provide a well-mixed aerosol directly downstream of the diluter. This allows taking a representative sample of the diluted aerosol for further conditioning or measurement by splitting the aerosol flow directly downstream of the diluter, and allows for a compact sampling system. The primary dilution stage is typically heated to 350 °C, while the second stage is operated at ambient temperature. The dilution factor of the system is approximately 80. The exact value is dependent on the inlet flow and the mass flow management: The flow rates in the sampling system are managed by a system of two mass flow controllers and two mass flow meters. The mass flow controllers control the dilution air flow rates. The mass flow meters monitor the flow rates extracted downstream of dilution stages 1 and 2. The differences between the flows extracted and the flows supplied can be changed. In other words, the net flow added or subtracted in one dilution stage can be defined. The sample flow rate, Q_{sample} , is defined as the sum of all other flow rates: 1) Flow rate drawn by the measurement instruments (Q_{inst}); 2) the dilution air flow rates ($Q_{dil,i}$); and 3) the excess flow rates $Q_{ex,i}$. For the calculation of the sample flow, the contributions of the flows extracted from the system are positive and the contributions of the flows fed into the system are negative.

$$Q_{sample} = Q_{ex,1} + Q_{ex,2} + Q_{inst} - Q_{dil,1} - Q_{dil,2}$$

The total dilution ratio DR is calculated by:

$$DR = DR_1 \times DR_2 = \left(\frac{Q_{dil,1}}{Q_{sample}} + 1 \right) \times \left(\frac{Q_{dil,2}}{Q_{sample} + Q_{dil,1} - Q_{ex,1}} + 1 \right)$$

A catalytic stripper (CS) is situated between dilution stage 1 and 2 and is operated at 350 °C at a flow rate of 1 liter per minute (L/min). The catalytic stripper provides oxidation of organic compounds and sulphur storage. The removal of these substances ensures the isolation of the solid particle fraction. The undesired formation of volatile and semivolatile particles and growth of subcut size particles is prevented. The catalytic stripper used is commercially available (AVL GmbH). The volatile particle removal efficiency of the CS was verified with polydisperse emery oil particles >50 nm and >1 mg/m³ (3.5–5.5 mg/m³) showing an efficiency of >99% (actual value 99.9%) as defined by RDE regulations²⁰. This is a more rigorous test than the tetracontane test prescribed in the current PMP protocol.

One or more condensation particle counters are used to measure the particle number concentration downstream of the second dilution stage. A CPC with a d_{50} of 23 nm enables the measurement of the currently regulated emission of solid particles larger than 23 nm. Additionally, measuring the particle number concentration with one or more CPCs with a lower d_{50} cut point (e.g., 10 nm, 4 nm) enables the assessment of the currently unregulated solid particle fraction <23 nm down to the d_{50} cut size of the applied CPC.

The dilution air supply line, the primary porous tube diluter, and the catalytic stripper have independent heating elements containing k-type thermocouples (TC). Independently heating

different sections controls the temperature distribution in the system.

In addition to the thermocouples in the heating elements, two thermocouples are placed downstream of dilution stage 1 and 2. These two thermocouples directly measure the aerosol temperature.

Two absolute pressure sensors (NXP MPX5100AP) are used to monitor the pressure at the inlet and the outlet of the sampling system.

For mobile measurements, a Clayton Power LPS 1500 battery pack is used. A 10 L synthetic air bottle supplies the system with dilution air during mobile applications. The sizes of the battery and the gas bottle are chosen so that the system can operate independently for 100 min.

The system is controlled via a NI myRIO running a LabVIEW virtual instrument. The virtual instrument allows for control of the flow rates and heater temperatures. Apart from the controlled parameters, the aerosol temperatures, pressures, and acceleration (via the sensor integrated in myRIO) can be monitored and logged. A myRIO accessory GPS module enables logging of the position data. **Figure 3** and **Figure 4** show the user interface of the virtual instrument used for controlling the DTT system.

[Place **Figure 3** here]

[Place **Figure 4** here]

Any kind of sampling procedure causes particle losses. To be able to account for these losses, laboratory measurements are performed to determine the particle size dependent particle penetration through the DTT sampling system. In these measurements, the particle concentration of monodisperse aerosol is measured upstream and downstream of the sampling system using two condensation particle counters. **Figure 5** shows the experimental setup for the calibration measurements. In this setup, a Jing miniCAST is used as a particle source^{21,22}. Mass flow controllers (MFC) are used to control the gas flows into the burner. A dilution bridge enables the adjustment of the particle number concentration. The dilution bridge is a high-efficiency particulate air (HEPA) filter parallel to a needle valve. Adjusting the position of the needle valve alters the dilution ratio by changing the ratio between the fraction of the aerosol passing through the HEPA filter and the fraction of the aerosol passing through the needle valve. The filtered and the unfiltered aerosols are recombined with a T-piece to form a diluted aerosol. A catalytic stripper is used to remove possibly abundant volatile compounds generated as byproducts of the combustion process. A TSI 3082 electrostatic classifier together with a TSI 3085 differential mobility analyzer (nano DMA) are used for the size selection of particles. Two TSI CPCs 3775 ($d_{50} = 4$ nm) are used to measure the particle number concentration upstream and downstream of the DTT sampling system. The counters' cut point of $d_{50} = 4$ nm allows for the penetration determination at particle sizes as low as 10 nm and below.

[Place **Figure 5** here]

PROTOCOL:

1. Calibration procedure

1.1. Set up and prepare instruments.

1.1.1. Place the instruments described, shown in **Figure 5**, in an organized and compact way in a laboratory with an extraction system.

1.1.2. Connect the instruments as indicated by the arrows in **Figure 5** using conductive tubing. Keep the tubing as short as possible to minimize diffusional particle losses.

1.1.3. Connect the instruments requiring power (i.e., DTT system, DTT system pump, two CPCs, DMA, catalytic stripper, and MFCs) to sockets.

1.1.4. Connect the CPCs, the DTT system, and the MFC to a laptop.

1.1.5. Make sure the laptop has the required software installed to communicate with the connected devices.

1.1.6. Install missing software if required.

1.2. Warm up experimental components at least 30 minutes before starting the calibration measurements to ensure a thermally stable measurement setup.

1.2.1. Start the operation of the burner by setting the gas flow controlled by the external MFC to the start setting specified in the user's manual.

1.2.2. Ignite the flame.

1.2.3. Feed the generated soot into the extraction system.

1.2.4. Produce soot particles with a mean diameter of 50 ± 5 nm by setting the MFC-controlled flows accordingly. A table of settings and expected particle size distribution can be found in the burner manual or in the literature²³. For the miniCAST settings in **Table 2** can be used:

[Place **Table 2** here]

1.2.5. Start heating the catalytic stripper by setting the corresponding temperature controller to 350 °C.

1.2.6. Switch on the CPCs and set to low flow mode (i.e., inlet flow of 0.3 L/min).

1.2.7. Set up the communication of the CPCs with the laptop using the CPCs manufacturer's software or serial communication.

1.2.8. Start up the DTT system warm up procedure as described in section 3.1.

1.2.9. Install the impactor with a 0.071 cm nozzle at the inlet of the classifier according to the user's manual.

1.2.10. Switch on the classifier. The display on the classifier should show an impactor flow of 1.30 ± 0.05 L/min. If the flow shown is different, double check the tubing connecting the classifier with the CPC and the DTT system.

1.2.11. Set the sheath flow rate of the classifier to 13 L/min using the user interface.

1.2.12. If a soft X-ray source (TSI 3088) is used, switch on the classifier's neutralizer.

1.3. After at least 30 min of warm up time perform the calibration measurements.

1.3.1. Stop feeding the generated soot into the extraction system and connect the outlet of the burner to the dilution bridge.

1.3.2. Set the particle size selected by the classifier to 10 nm using the user interface.

1.3.3. Using the dilution bridge needle valve, adjust the particle number concentration upstream of the DTT system to be $10^4 \pm 10^3$ #/cm³. This particle concentration yields a relatively high signal, enabling short measurement times while the CPCs operate in single-count mode, which ensures high accuracy. If the desired concentration of $10^4 \pm 10^3$ #/cm³ cannot be reached because of exceedingly low particle concentrations emitted by the soot generator, maximize the throughput through the dilution bridge by fully opening the valve.

1.3.4. Start logging the data of the DTT system (if not started already) by clicking the "**Start Data Logging**" button in the DTT Labview software.

1.3.5. Start logging the data of the two CPCs using the proprietary software or serial communication.

1.3.6. Wait 30 s for the experimental setup to stabilize.

1.3.7. Note down a timestamp and the set particle size to mark the start of the measurement.

1.3.8. Run the measurement for 2 min.

1.3.9. Note down a timestamp to mark the end of the measurement.

1.3.10. Repeat steps 1.3.3–1.3.9 for particle sizes of 15 nm, 30 nm, 50 nm, and 100 nm. Additional measurements can be taken if better size resolution is desired.

1.3.11. Perform another set of measurements at the same particle sizes as before by repeating steps 1.3.2–1.3.10.

1.3.12. Stop logging the measurement data of the two CPCs and the DTT system.

1.3.13. Shut down all the instruments.

1.4. Evaluate the collected calibration data with a spreadsheet program.

1.4.1. Export the particle concentration data measured by the CPCs into a .csv or .txt file.

1.4.2. Import the CPC and the DTT system data into a data evaluation tool.

1.4.3. Assign the data to the corresponding measurements by allocating data from each instrument (i.e., 2 CPCs, DTT system) with a timestamp between the start and the end timestamp of a measurement to the corresponding measurement. It is recommended to automate this task with a data evaluation tool.

1.4.4. Time average the two particle concentration datasets (CPCs) and the dilution ratio (DTT system) for all measurement points.

1.4.5. Calculate the relative particle penetration for all measurement points according to the following formula:

$$P_n = \frac{\overline{C_{d,n}}}{\overline{C_{u,n}}} \overline{DR_n}$$

Where P_n is the relative particle penetration at a certain measurement point n . $\overline{C_{d,n}}$ is the particle concentration measured by the CPC downstream of the DTT system averaged over the timespan of the measurement point n . $\overline{C_{u,n}}$ is the corresponding particle concentration measured by the CPC upstream of the DTT system averaged over the timespan of the measurement point n . $\overline{DR_n}$ is the dilution ratio from the DTT system, averaged over the timespan of the measurement point n .

1.4.6. Calculate the mean particle penetration P_{mean} by averaging over the average particle penetrations at 30 nm, 50 nm, and 100 nm particle size.

$$P_{mean} = \frac{P_{30nm} + P_{50nm} + P_{100nm}}{3}$$

This value is used for the calculation of the Particle Concentration Reduction Factor (PCRF)

dividing the dilution ratio DR with the mean penetration efficiency P_{mean} .

$$PCRF = \frac{DR}{P_{mean}}$$

The $PCRF$ is calculated from the penetration at 30 nm, 50 nm, and 100 nm to be comparable with PMP compliant, commercially available instruments. The measurements at sizes other than 30 nm, 50 nm, and 100 nm are used to determine the d_{50} cutoff size of the system to better characterize the system outside the regulatory frame.

2. Installation and preparation for real driving emissions measurements

2.1. Select a vehicle to assess for particle number emissions for particles <23 nm.

2.2. Select a route to measure the particle number emissions of the selected vehicle. There are guides on how to select appropriate routes in the literature²⁴.

2.3. Exhaust flow meter (EFM) installation

2.3.1. Choose an EFM with a measurement range matching the expected exhaust flow range of the vehicle to be measured²⁴.

2.3.2. Place the EFM control box in the trunk of the vehicle.

2.3.3. Install the EFM outside of the car, according to the manufacturer's specification sheet. **Figure 6** shows an example of an installed EFM, mounted externally on shaped pipes leading into the trunk.

2.3.4. Make sure the distance upstream and downstream of the EFM comply with EU regulations (i.e., 4x the pipe diameter or 150 mm straight pipe, whichever is larger, should be upstream and downstream of the flow sensor).

2.3.5. When measuring vehicles with multiple exhaust manifolds, the individual exhaust pipes should be joined in front of the EFM and the cross-sectional area of this pipe increased accordingly to keep the increase in exhaust backpressure as low as possible. If this is not possible, the exhaust mass flow can be measured with several EFMs.

2.3.6. Make sure the connectors from the EFM pipe to the exhaust pipe of the vehicle can withstand the exhaust gas temperatures (i.e., no plastic should be used).

2.3.7. The pipe diameter, the connector diameter, and the diameter of any extensions required for sampling should not be smaller than the diameter of the exhaust pipe to keep the exhaust back pressure as low as possible.

2.3.8. Start the piping at the exhaust of the vehicle.

2.3.9. Connect the exhaust to the first pipe with connecting pipes and pipe clamps. Tighten the pipe clamps only at the end in order to be able to align the pipes during fitting.

2.3.10. Connect one pipe at a time with connecting pipes and pipe clamps until there is a connection from the exhaust to the EFM. This should be as short as possible.

2.3.11. Place the EFM control box and the EFM mounting bracket in the trunk to ensure that nothing slips during the measurement trip.

2.3.12. Check that all the piping is tight and nothing comes loose during the measurement trip.

2.3.13. Switch on the EFM.

2.3.14. After a warm up time of up to 15 min depending on ambient temperature (see EFM user guide), the exhaust mass flow meter is ready to measure^{25–28}.

[Place **Figure 6** here]

2.4. Preparing and installing the DTT measurement system in the trunk of the vehicle

NOTE: The measurements described here are conducted with two condensation particle counters as counting devices for the DTT system. One of the CPCs (TSI 3790A) has a lower d_{50} cutoff size of 23 nm, which equals the current legislative limit. The other CPC (commercially available 10 nm AVL CPC) has a lower d_{50} cutoff of 10 nm. Measuring particle emissions with these two instruments in parallel enables the assessment of the currently regulated emissions (>23 nm) and the <23 nm fraction.

2.4.1. Take a laptop and install the DTT software and the software for logging the CPC measurement data.

2.4.2. Place the synthetic air bottle in the trunk or on the floor in front of the rear seats and fix it using straps.

2.4.3. Place the battery in the trunk of the vehicle and fix it. Plug in the AC input cable and connect it to a local power source.

2.4.4. Place and fix the vacuum pumps for the sampling system and the condensation particle counters in the trunk of the vehicle and connect them to the battery.

2.4.5. Place the DTT system in the trunk of the vehicle and fix its position using straps. **Figure 7** and **Figure 8** show the DTT system in the trunk of a car. Connect the system to the mobile battery pack.

2.4.6. Connect the two inlet MFCs of the DTT system to a stationary pressurized air supply. Connect the two outlet MFMs of the DTT system to the vacuum pump.

2.4.7. Use appropriate tubing to drive the exhaust of the pump outside the vehicle.

2.4.8. Connect the DTT system to the measurement laptop using a USB cable.

2.4.9. Connect the inlet of the system to the sampling point downstream of the EFM. Connect the system power inlet to the battery. Connect the condensation particle counters' power inlets to the battery pack.

2.4.10. Connect the CPCs to the respective external vacuum pump.

2.4.11. Mount the butanol bottles of the CPCs firmly on the frame of the dilution system as far away as possible from the vehicle occupants.

2.4.12. Make sure that the cap is screwed on tight and does not open during the measurement drive when accelerating.

2.4.13. Use appropriate tubing to drive the exhaust of the CPCs and/or the external pump outside the vehicle. Connect the CPCs to the measurement laptop using USB cables.

NOTE: **Figure 9** shows the prepared vehicle. The DTT system is installed in the trunk of the vehicle. A commercially available PN-PEMS system is also installed to use as a reference for the regulated emission of solid particles >23 nm.

[Place **Figure 7** here]

[Place **Figure 8** here]

[Place **Figure 9** here]

3. Measurement operation

3.1. Heating and starting up the measurement system

3.1.1. Switch on the two CPCs and their external vacuum supply.

3.1.2. Open the CPCs software on the measurement laptop and establish communication with the CPCs. The communication can either run via the instrument's proprietary software or via serial communication as described in the CPC manual.

3.1.3. Close the needle valves downstream of the MFMs.

3.1.4. Switch on the DTT sampling system pump.

3.1.5. Switch on the sampling system by pushing the red switch down.

3.1.6. Open the LabVIEW DTT application on the computer. The communication with the system starts automatically.

3.1.7. The graphical user interface (GUI) of the DTT LabVIEW application now displays the flows in and out at dilution stages 1 and 2, which should be 0.00 L/min. If not, double check that the needle valves are closed properly.

3.1.8. Enter the mass flow drawn by the connected measurement instruments in sL/min. If the flow drawn by the instruments is unknown, measure it using a handheld mass flow meter (e.g., Vögtlin red-y compact series). Reconnect the tubing after measuring the flows drawn by the CPCs.

3.1.9. Slowly open the needle valves until both "Flows out" reach 10.0 ± 0.5 sL/min. Both "Flows in" will increase to the same values as the corresponding "Flows out".

3.1.10. Adjust the "Add Flow" (i.e., difference between dilution air flow and excess flow) of both dilution stages to get $Q_{CS} = 1.0 \pm 0.1$ L/min through the catalytic stripper and a sample inlet flow of $Q_{sample} = 1.0 \pm 0.1$ L/min.

3.1.11. Click on the "Heater" tab to set the heater temperatures.

3.1.12. Set the heater temperatures of the dilution air supply, the first porous tube diluter, and the catalytic stripper to 350 °C. The system will now start to heat up. Below the "Set" interfaces the current temperature and heating power percentages are displayed.

3.1.13. Wait until the gas temperature downstream dilution stage 1 ("T DilStage 1" in the GUI) reaches 290 °C before starting the measurement drive. This will take approximately 20 min.

3.2. Data logging

3.2.1. Start to log the data on the measurement devices connected to the DTT sampling system.

3.2.2. Start to log the data of the sampling system by pressing the "Start Data Logging" button and choose a path and a file name in the pop-up window. The log file path will be displayed and the green light will indicate that data are saved. The system data are logged at a frequency of 2 Hz.

3.2.3. Log the particle concentration data of the CPC using appropriate software. This can either be the manufacturer's or a serial communication software (e.g., PuTTY).

3.2.4. Start logging the exhaust flow with the EFM.

3.3. Driving

3.3.1. Before driving the selected route, disconnect the battery's charging cable and switch from stationary pressurized air supply to the gas bottle.

3.3.2. Drive the selected route.

3.4. After driving

3.4.1. Press "Logging ..." to stop recording data. Shut down the instruments.

3.5. Recharge the battery to prepare for the next drive.

4. Data analysis

4.1. Import the data from the sampling system, the vehicle (for exhaust flow), and the measurement devices into the same data analysis program.

4.2. Perform the time alignment considering the time the exhaust needs to be transported from the tailpipe to the measurement devices. The transport time t_{dil} through the dilution system is 2.5 s. The transport time t_{sample} through the sampling line can be calculated as follows:

$$t_{sample} = \frac{A_{sample} * l_{sample}}{\dot{Q}_{sample}}$$

Where t_{sample} is the transport time through the sampling line in seconds, t_{dil} is the transport time through the dilution system (2.5 s), A_{sample} is the cross section area of the sampling line in m², l_{sample} is the length of the sampling line from sample point to the dilution system inlet in meters, and \dot{Q}_{sample} is the DTT Dilution System sample flow in m³/s. Add t_{sample} to t_{dil} to get the total delay time t_{total} :

$$t_{total} = t_{sample} + t_{dil}$$

NOTE: As an example, t_{total} for a pipe length of 0.5 m with an internal pipe diameter of 4 mm and sample flow of 1 L/min equals 2.88 s. **Figure 10** shows an example of the time alignment of the measured particle number (blue dotted line) to the time shifted particle number (blue line).

[Place **Figure 10** here]

4.3. To be able to calculate the particle number \dot{PN} in #/s, the exhaust gas volume flow $\dot{V}_{exhaust_norm}$ in cm³/s must be calculated first according to the following formula:

$$\dot{V}_{exhaust_norm} = \frac{\dot{m}_{exhaust} * R * T_{norm}}{p_{norm}}$$

where $\dot{V}_{exhaust_norm}$ is the exhaust standard volume flow in m³/s, $\dot{m}_{exhaust}$ is the measured exhaust mass flow in kg/s, R is the ideal gas constant for air (287.1 J/kg*K), T_{norm} is the temperature at standard conditions (273.15 K), and p_{norm} is the pressure at standard conditions (101,330 Pa). With this exhaust volume flow at standard conditions the particle number can be calculated by multiplying the $\dot{V}_{exhaust_norm}$ with the dilution ratio DR of the sampling system, the concentration c_{PN} measured by the CPCs, and the factor 10⁶ (for the conversion from m³ to cm³).

$$\dot{PN} = \dot{V}_{exhaust_norm} * c_{PN} * DR * 10^6$$

4.4. To correct for particle losses, multiply the particle exhaust flow times the particle number concentration rate with the system particle concentration reduction factor ($PCRF$) instead of the dilution ratio DR . The determination of $PCRF$ is described in the calibration instruction section 0:

$$\dot{PN}_{corr} = \dot{V}_{exhaust_norm} * c_{PN} * PCRF$$

REPRESENTATIVE RESULTS:

Calibration Data (Particle Penetration):

Figure 11 shows an exemplary plot of the relative particle penetration of the DTT system as a function of the particle mobility diameter. The corresponding data have been measured and evaluated as described in instruction point 0. The plot shows that the deviations between two measurement points at the same mobility diameter were less than 5%. Deviations larger than 10% indicate instabilities in the experimental setup. In this case, the calibration had to be repeated with increased warm up stabilization times. Both the warm up time (typically 30 min) and the stabilization time (typically 30 s) increased by a factor of 1.5.

The particles passing through the DTT system were lost due to diffusion and thermophoresis. Thermophoretic losses were caused by a temperature gradient drawing particles towards the walls of the sampling system. This is a particle size independent effect²⁹; in contrast, diffusion is highly particle size dependent. A concentration gradient caused a net particle flux towards the walls where particles were lost. The diffusivity rising with lower particle size made this the dominant loss mechanism for particles ≤10 nm. The lines in **Figure 11** indicating thermophoretic, diffusional, and total losses demonstrate the respective particle size dependencies. For the diffusional losses, this function was used to illustrate the approximate particle size dependency:

$$P = \exp\left(-\frac{a}{D(d_p)}\right)$$

The penetration P depends on a fit parameter a and the diffusion coefficient D :

$$D = \frac{kTC_c}{3\pi\eta d_p}$$

The diffusion coefficient depends on the Boltzmann constant k , the absolute temperature T , the viscosity η , the particle diameter d_p , and the Cunningham slip correction factor C_c , which is a function of the mean free path and the particle diameter²⁹.

The data illustrated in **Figure 11** resulted in the following mean particle penetration efficiency P_{mean} :

$$P_{\text{mean}} = \frac{P_{30\text{nm}} + P_{50\text{nm}} + P_{100\text{nm}}}{3} = \frac{0.795 + 0.825 + 0.825}{3} = \underline{\underline{0.815}}$$

The particle size where the penetration efficiency amounts to 50% is referred to as d_{50} . The d_{50} describes the penetration cutoff characteristic of a system. For the DTT system the d_{50} was 11 nm. The d_{50} is shown in **Figure 11**.

[Place **Figure 11** here]

Solid Particle Number:

Figure 12 shows the particle number emission rate over time for the first ten minutes of an RDE measurement drive. The data from the DTT PEMS using a 10 nm and a 23 nm CPC are shown together with data from a commercially available 23 nm cut point system. The particle emission rates were calculated from the respective particle concentrations multiplied by the exhaust flow rate as described above in the data analysis instruction section 0. The reference instrument (AVL MOVE) relied on a diffusion charger for the particle number concentration measurement. Despite the different sensor principles, the data measured with the DTT PEMS were overall in very good agreement with the data measured by the commercially available PEMS. Sharp downwards pointing spikes in all three signals occurred because the particle measurement devices can report zero particle concentrations temporarily and zeros cannot be displayed in logarithmic plots. The particle emissions measured with the 10 nm CPC were very close to the emissions measured with the 23 nm CPC for the majority of the time period shown in **Figure 12**. However, right at the beginning between 10 s and 25 s there was an occurrence of significant <23 nm particle emission. The DTT 10 nm signal was significantly higher than the 23 nm signal of the DTT system and the AVL MOVE. In this case, >50% of the total number of particles emitted were between 10 nm and 23 nm. Cold start dynamic processes in non-thermal equilibrium can cause particle size distributions to differ from emissions from a hot vehicle³⁰. The discussion of these complex processes is beyond the scope of this work. Further information on this topic can be found in the literature^{31–33}.

[Place **Figure 12** here]

FIGURE AND TABLE LEGENDS:

Figure 1: Schematic drawing of the DTT particle number portable emission measurement system.

Figure 2: Top view picture of the DTT sampling system.

Figure 3: DTT virtual instrument dilution stage parameter overview.

Figure 4: DTT virtual instrument heater control panel.

Figure 5: Schematic drawing of the experimental setup used for the calibration of the DTT sampling system.

Figure 6: Picture of an installed EFM.

Figure 7: DTT PEMS from inside the vehicle.

Figure 8: DTT PEMS inside the trunk of a vehicle.

Figure 9: Vehicle with commercially available PN-PEMS (AVL MOVE) and DTT PEMS installed.

Figure 10: Example of time alignment of measured particle number PN in $\#/cm^3$ compared to the measured exhaust mass flow in kg/h.

Figure 11: Particle penetration as a function of particle mobility diameter. Points marked in blue are measurement results. The dashed lines in orange and green indicate the losses associated with thermophoresis and diffusion, respectively. The red line represents the total losses as the sum of diffusional and thermophoretic losses. The dotdashed purple line shows the average particle penetration P_{mean} as calculated in the calibration measurement instruction section 1.

Figure 12: The upper part of the figure shows the particle number emission rate over time for the first 10 mins of an RDE measurement drive. Data measured with the DTT PEMS using 10 nm and 23 nm CPC and a commercially available 23 nm cut point system (AVL MOVE) are used as a reference. The lower part of the figure shows the velocity of the vehicle.

Table 1: Elements of the PMP Measurement System.

Table 2: Suggested miniCAST flow rates for calibration measurements.

DISCUSSION:

This work presents the DTT sampling system and its application as a portable emission measurement system. The system was designed and constructed within the EU Horizon 2020 project DTT to enable particle number emission measurements below the current legislative particle size limit of 23 nm. The system's versatility enables the assessment of the regulated solid particle number emissions as well as total particle emissions and studies on secondary aerosols.

To interpret measurement results accurately, a calibration procedure is necessary with the DTT system. This is to evaluate the relative particle penetration for different particle sizes, to be able to calculate a correction factor that accounts for the particle losses. It is critical to provide sufficient warm up time for the sampling system itself and the rest of the experimental setup to reach thermal equilibrium and achieve accurate calibration measurement results.

The application of the DTT system for the measurement of solid particle number emissions with a lower particle size cutoff of 23 nm (current regulation) and 10 nm (experimental) is described. To be able to assess particle number emissions of a vehicle it is necessary to determine the particle number concentration and the exhaust mass flow rate. The DTT system covers the particle number concentration measurement. The exhaust mass flow is measured using an exhaust flow meter (EFM). It is critical to install the EFM according to the manufacturer's instructions. Erroneous measurements of the exhaust flow rate directly affect the deduced emission rates. When processing the measured data, it is important to perform an accurate time alignment of the particle concentration data and the exhaust flow data. This is necessary because the emission rate is the exhaust flow rate multiplied by the particle number concentration. If the two signals are not aligned correctly, the emissions over the whole drive can significantly deviate from the real emissions.

The DTT system is not a commercial device but a versatile research tool. It is used to investigate unregulated vehicle emissions as opposed to performing certification measurements validating compliance with current regulations. The high versatility comes at the cost of increased energy and dilution air consumption. When using the system for mobile measurements, the weight added to the vehicle due to the battery (30 kg) and gas bottle (20 kg) to cover the energy and air consumption of the system must be kept in mind. The total weight added to the car when measuring the PN emissions with the DTT system is approximately 80 kg, which is comparable to another person being transported in the vehicle. The added weight can lead to slightly increased emissions, especially if the drive includes a great deal of acceleration and/or hills.

The DTT system can be used to investigate the unregulated <23 nm particle number exhaust emissions. Both solid and total particle number emissions can be measured. Furthermore, it can be a useful tool to study the complex field of secondary aerosol formation. Another possible application of the system is the measurement of automotive brake wear particles. A significant fraction of the particles emitted during braking events can be smaller than 30 nm³⁴. With a d_{50} of approximately 11 nm, the DTT system is suitable for studying these emissions. Although it is known that non-exhaust emissions contribute almost equally to traffic-related PM₁₀ emissions³⁵, non-exhaust particle emissions are still unregulated. This is due to the complex and seldom reproducible process of particle generation, making it very difficult to set regulatory actions. Furthermore, the chemical composition and the related toxicity of organic brake wear particles is still widely unknown³⁵.

The DTT system is a useful tool to improve our understanding of both exhaust and non-exhaust traffic-related particle emissions.

ACKNOWLEDGMENTS:

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DISCLOSURES:

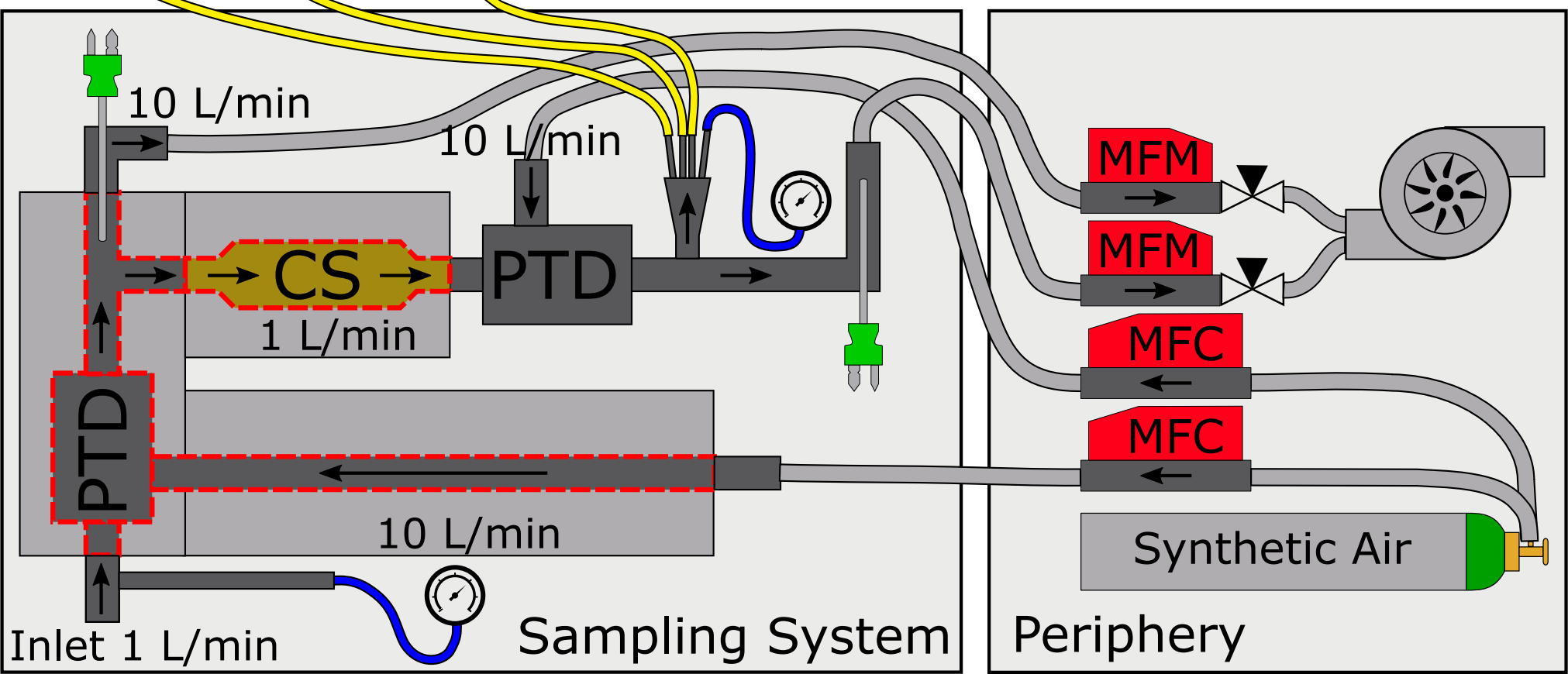
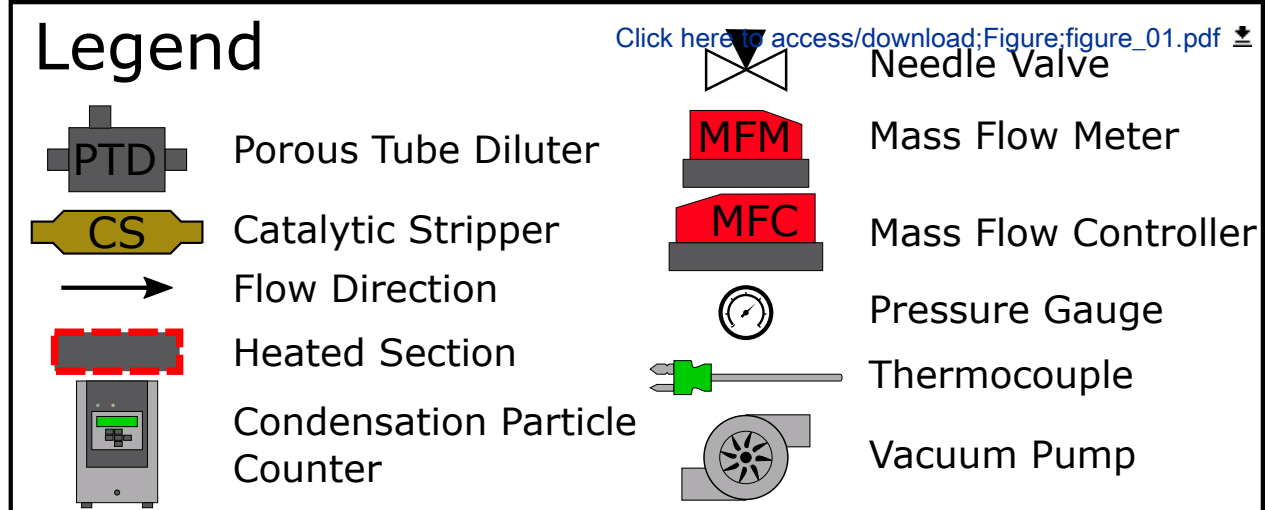
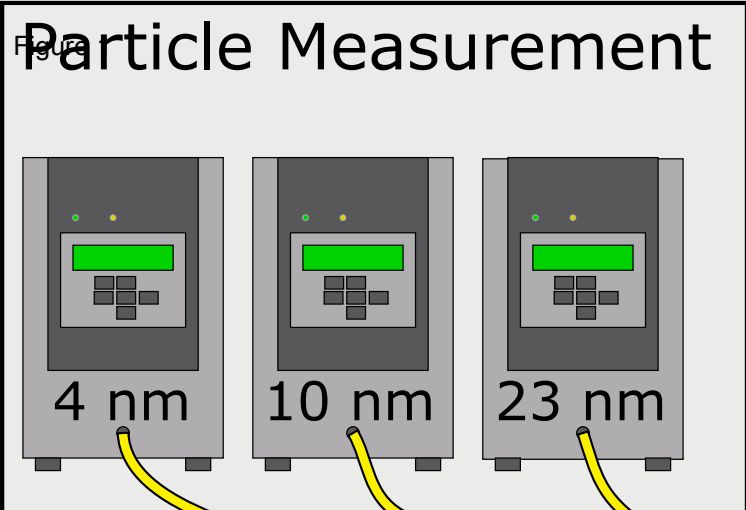
The authors have nothing to disclose.

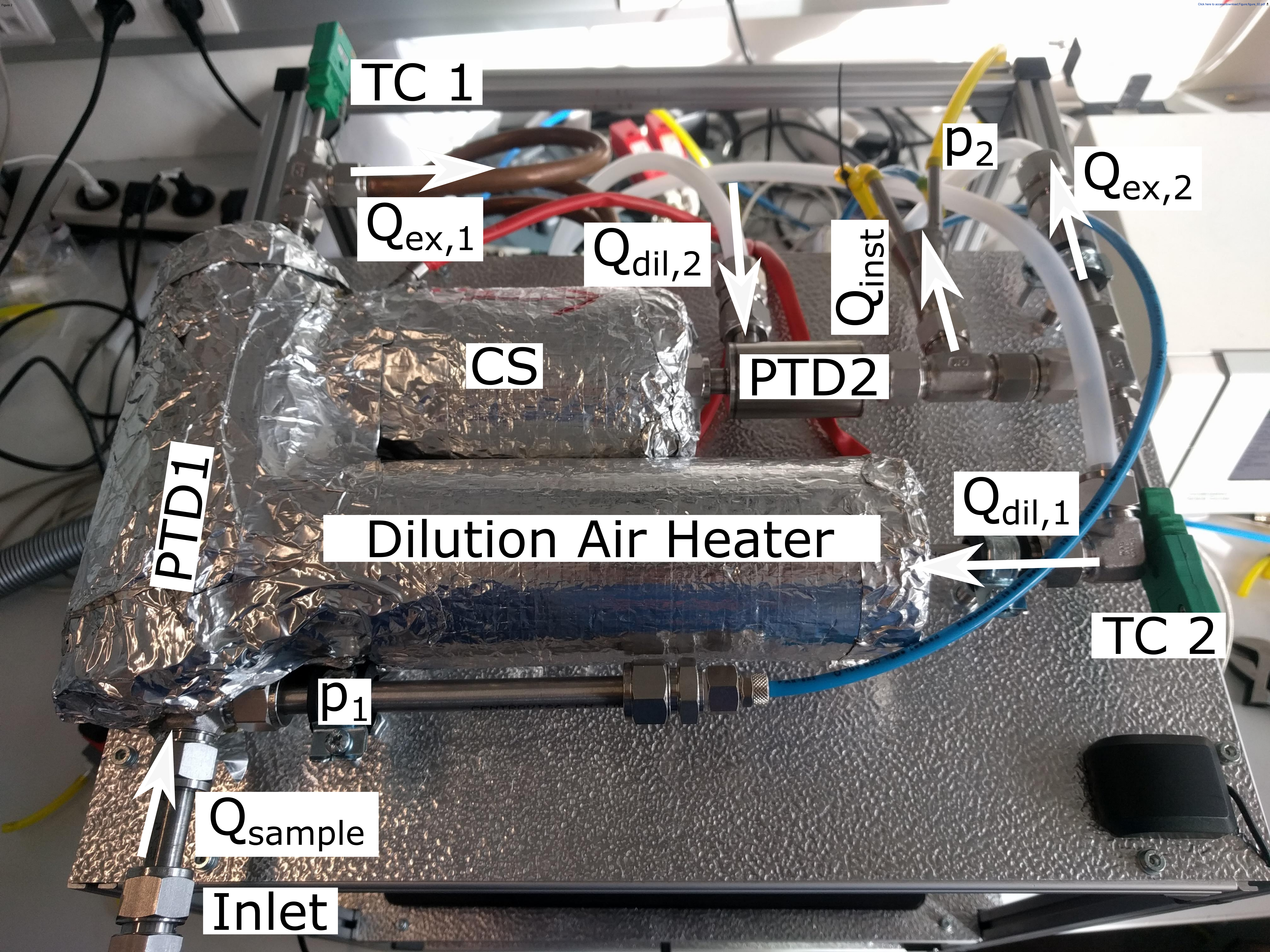
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888





TC 1

$Q_{ex,1}$

$Q_{dil,2}$

Q_{inst}

p_2

$Q_{ex,2}$

CS

PTD2

PTD1

Dilution Air Heater

$Q_{dil,1}$

TC 2

p_1

Q_{sample}

Inlet

Figure 3

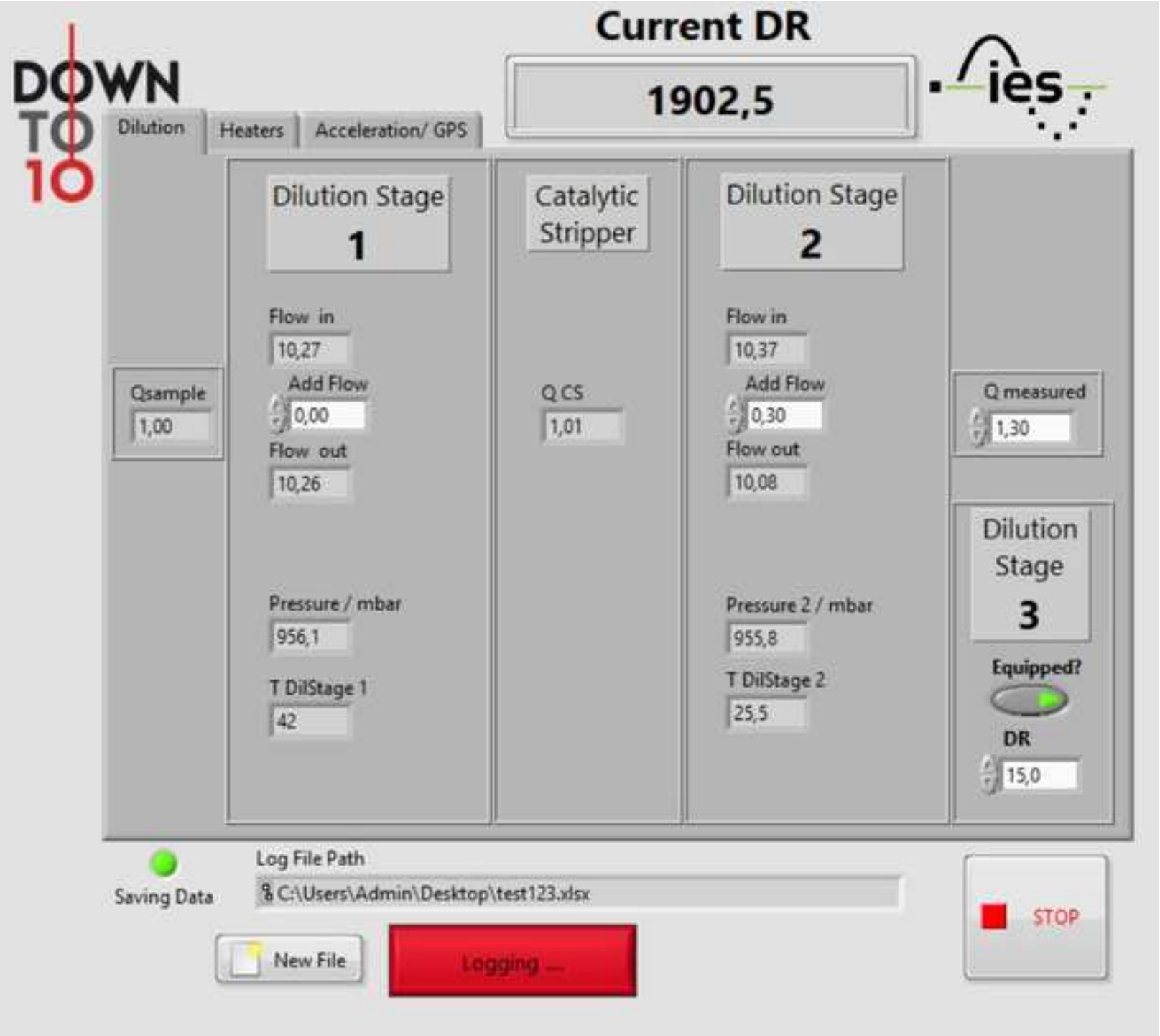
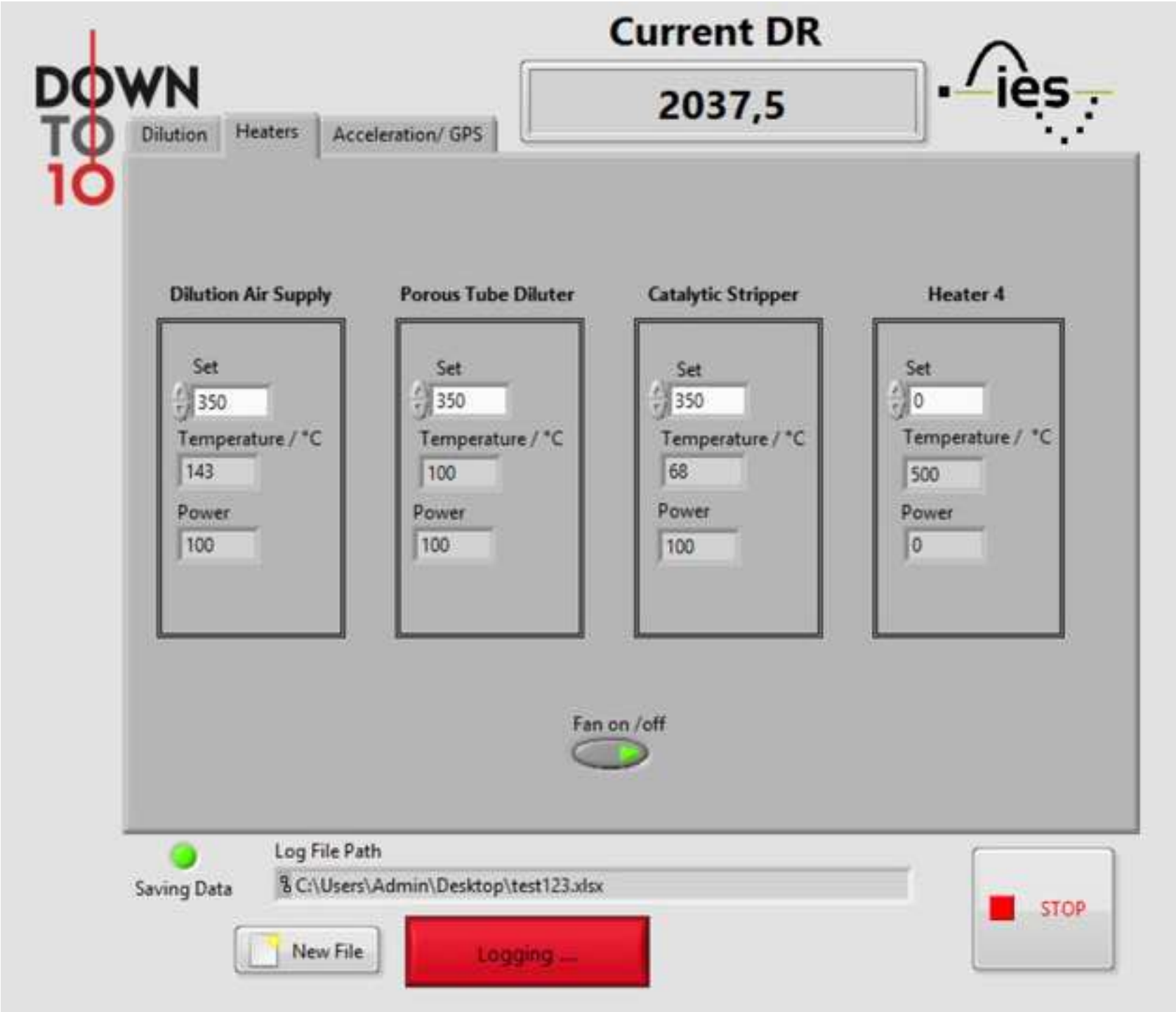


Figure 4



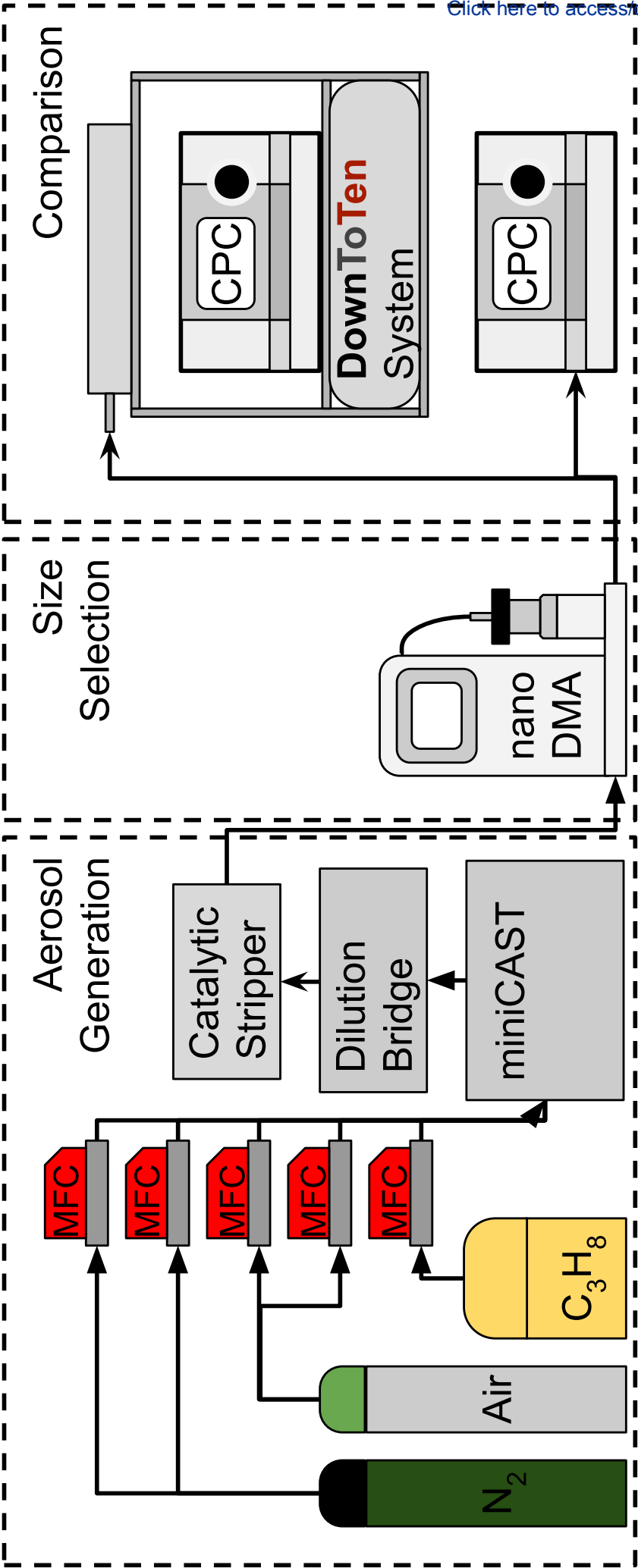
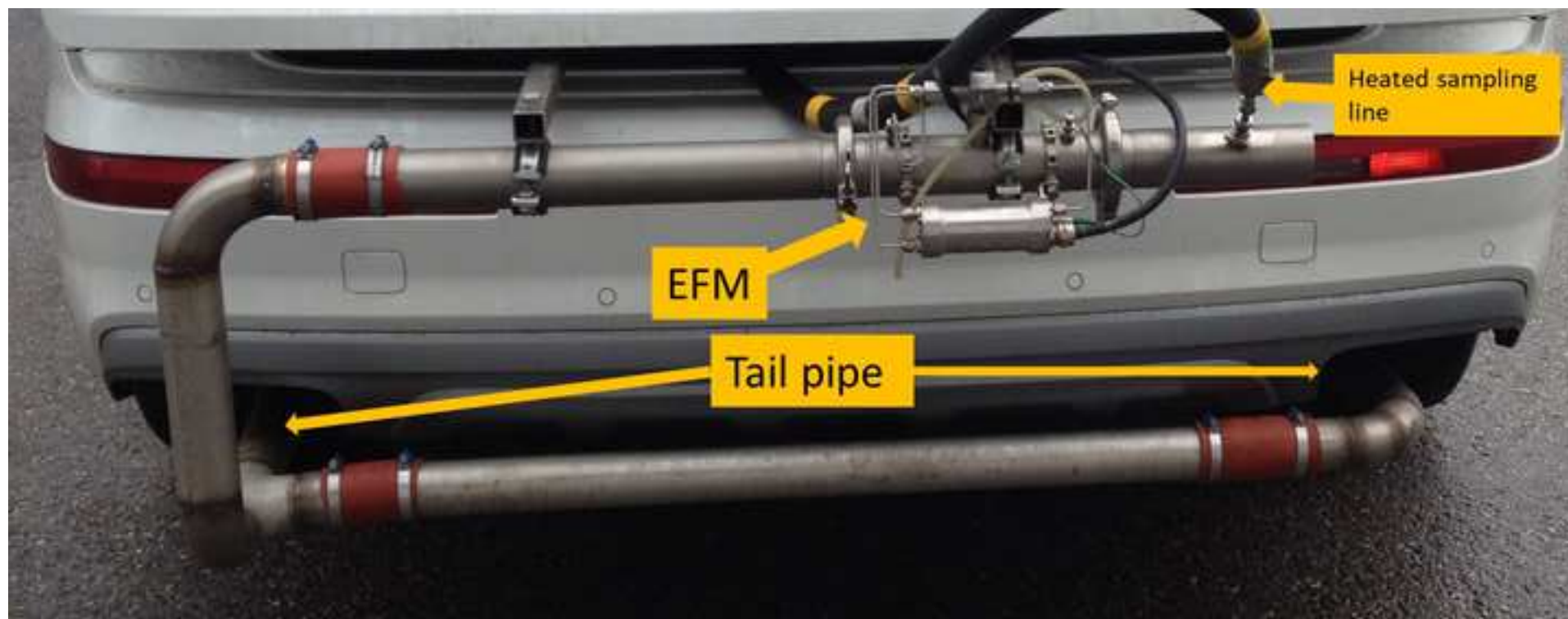
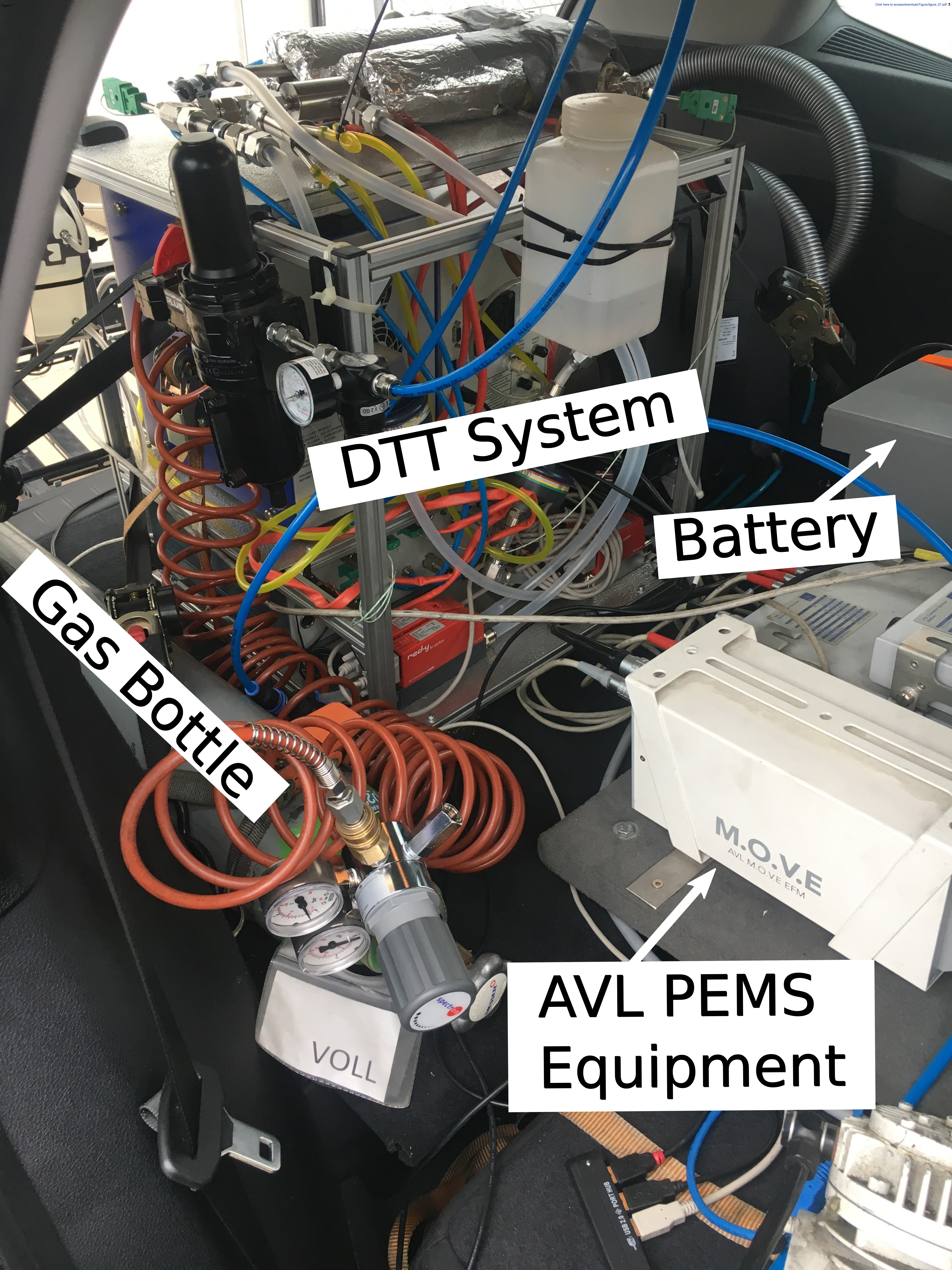


Figure 6





DTT System

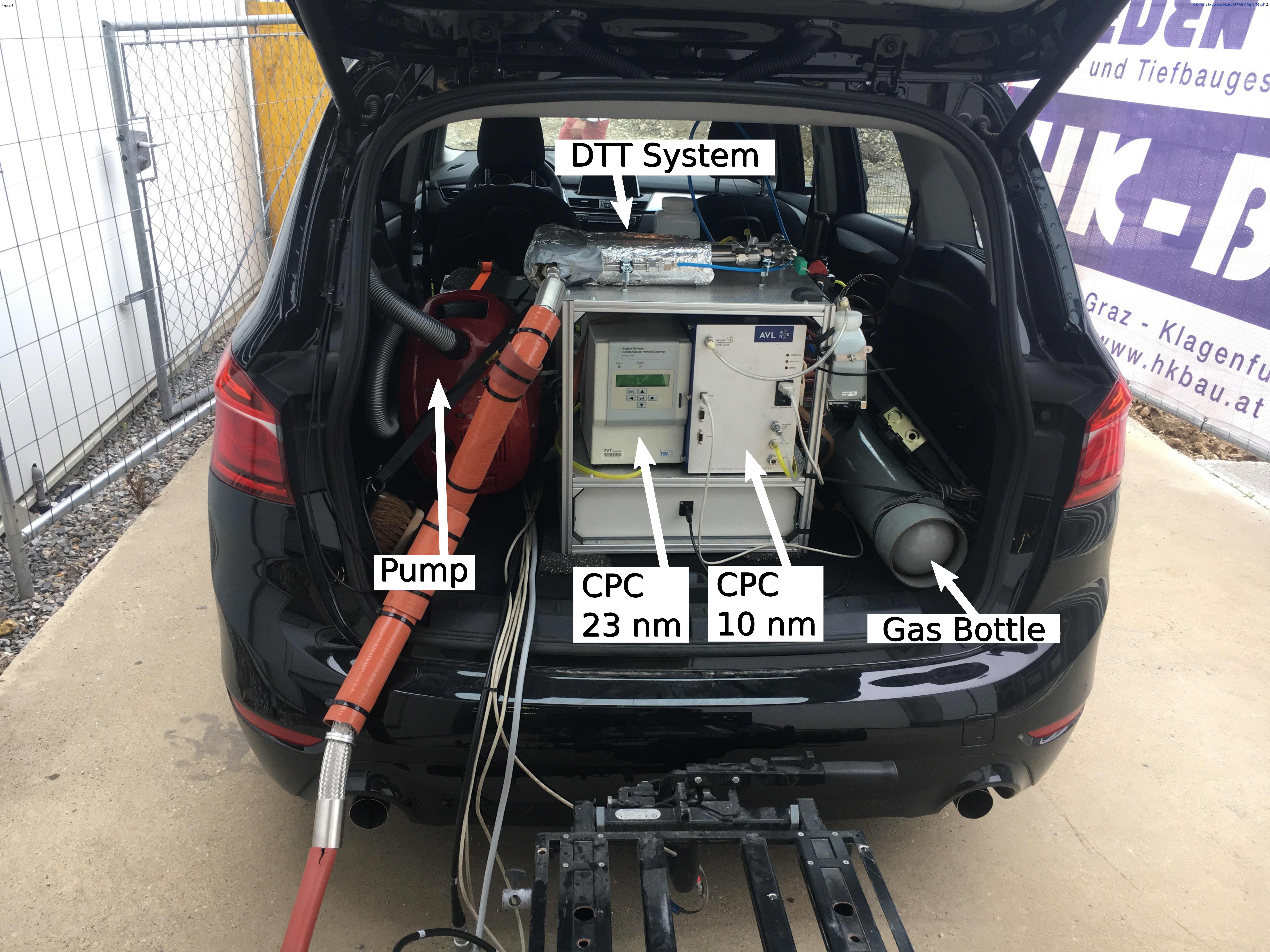
Battery

Gas Bottle

AVL PEMS
Equipment

M.O.V.E.
AVL M.O.V.E. EFM

VOLL



DTT System

Pump

CPC
23 nm

CPC
10 nm

Gas Bottle



MOTORPRÜFSTAND!
Türöffnungsbereich
freihalten!



218d



A

G



STEIERMARK

Figure 10

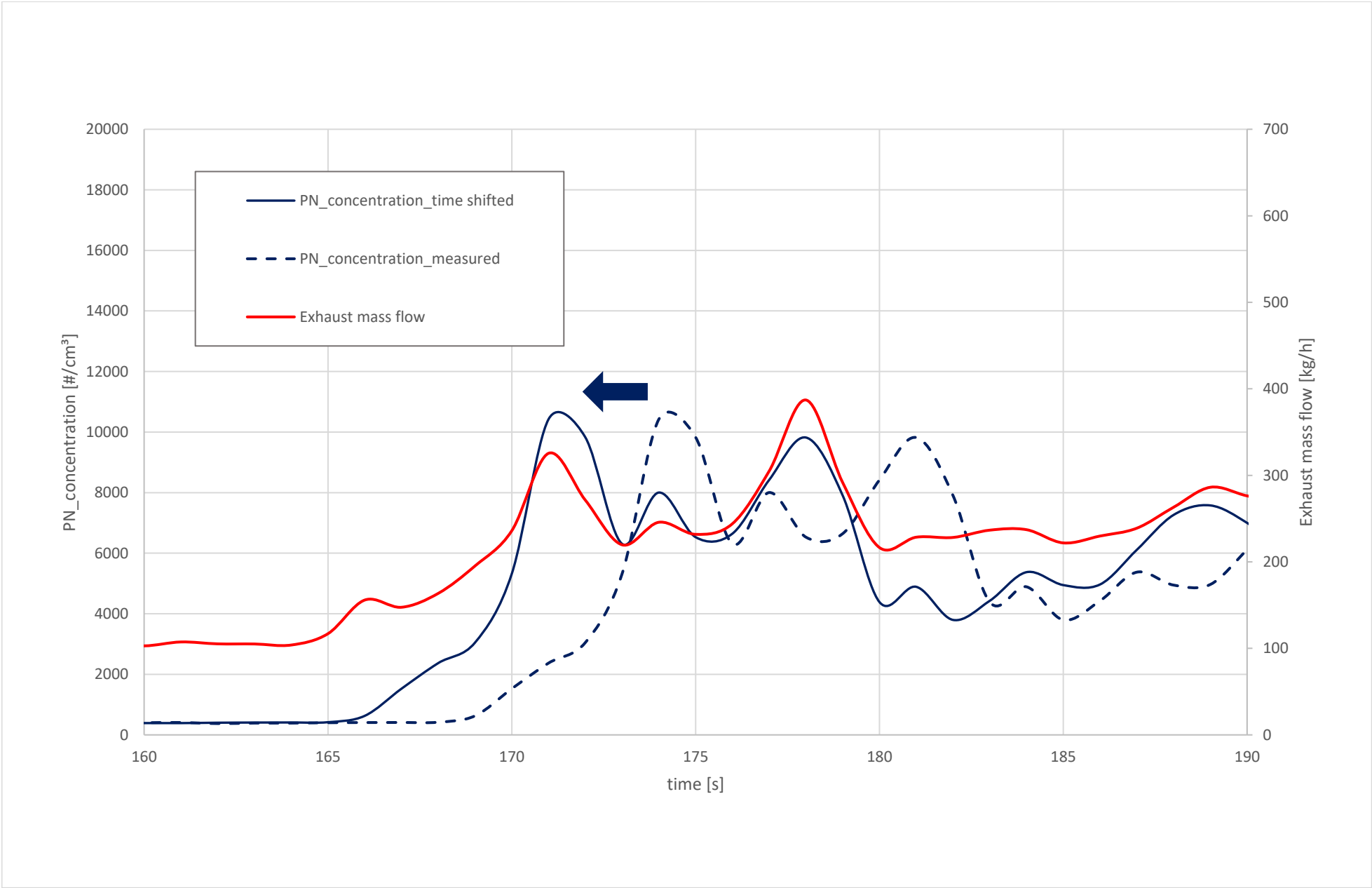
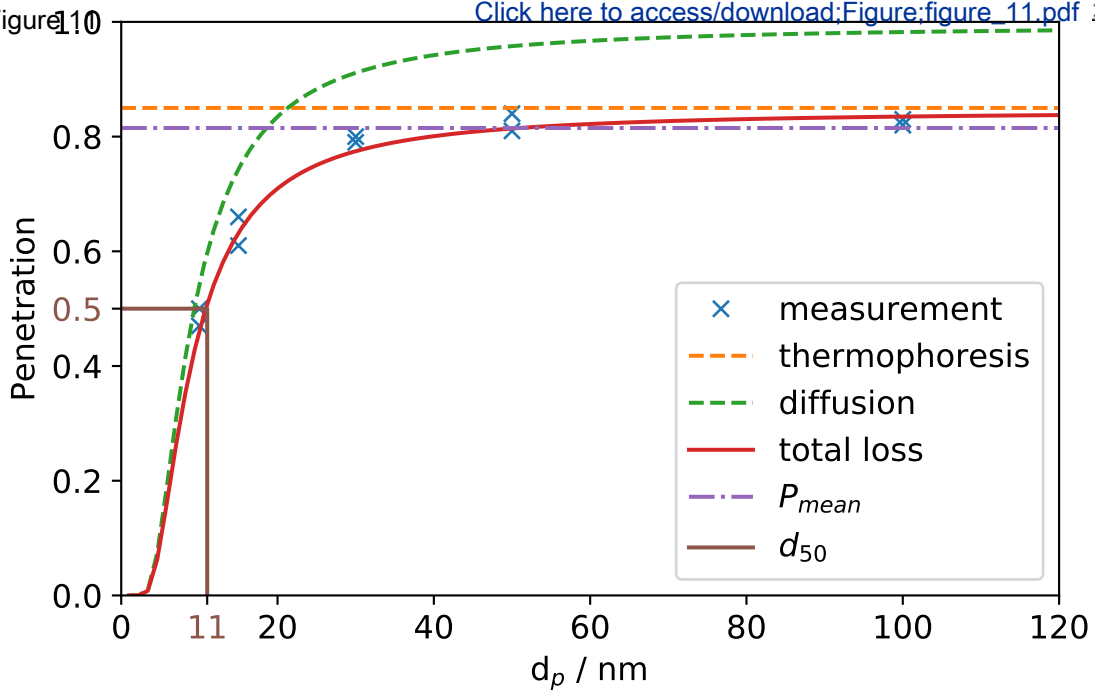
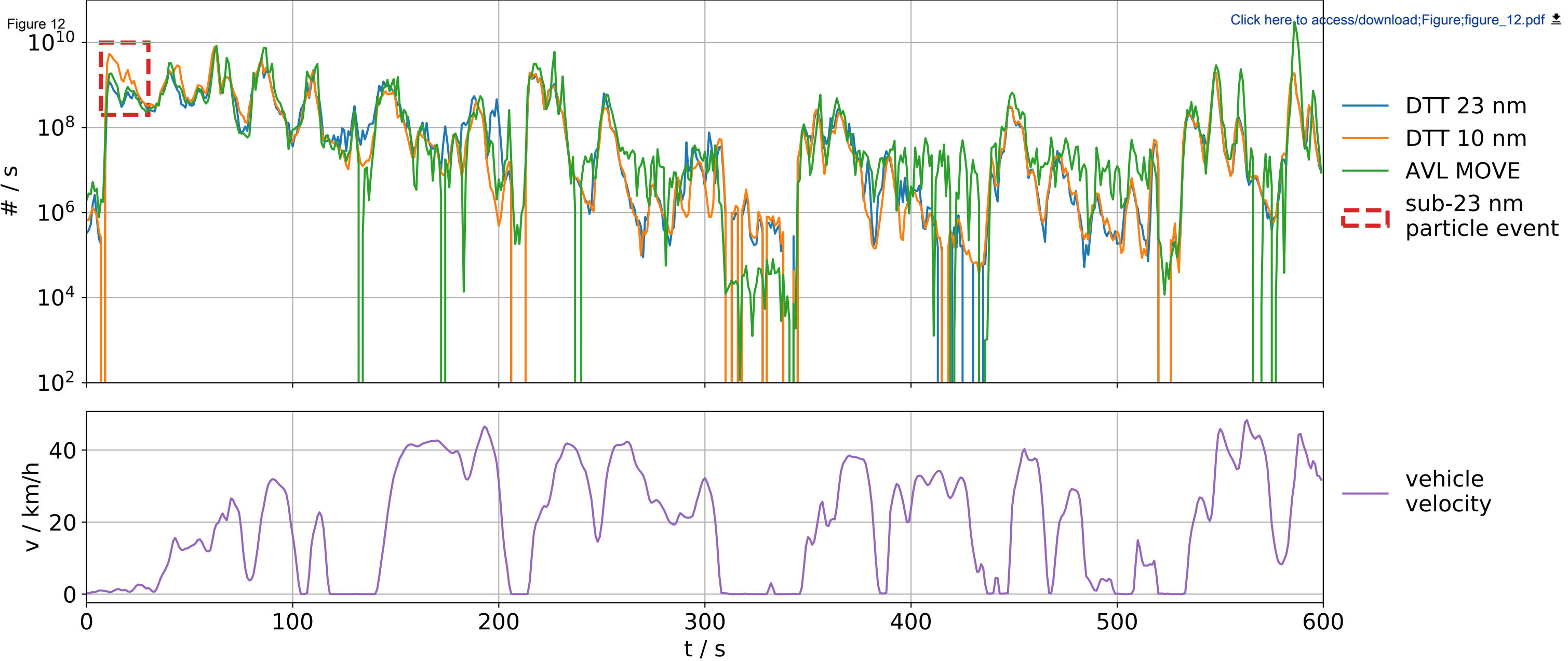


Figure 11





| Stage | Identity | Purpose |
|-------|---------------------------|--|
| 0 | Sample source | Origin of sample |
| 1 | Particle Transport | Conduct sample from origin to measurement system |
| 2 | Volatile Particle Remover | Eliminate volatiles and define non-volatile particles to be measured |
| 3 | Particle Number Counter | Enumerate non-volatile particles and define the lower size limit |

| Gas | Flow rate |
|------------------------------|-----------|
| Propane | 20 mL/min |
| Quench gas (N ₂) | 2 L/min |
| Dilution air | 5 L/min |
| Oxidation air | 0.5 L/min |
| Mixing gas (N ₂) | 0 L/min |

| Name of Material/ Equipment | Company | Catalog Number |
|---------------------------------------|---------------|----------------|
| 2x Condensation Particle Counter 4 nm | TSI | 3775 |
| 5x Mass Flow Controllers (MFC) | Vögtlin | |
| AVL M.O.V.E. EFM Exhaust Flow Meter | AVL | |
| Catalytic Stripper | Custom made | |
| Compressed Air | | |
| Condensation Particle Counter 10 nm | AVL | |
| Condensation Particle Counter 23 nm | TSI | 3790A |
| Differential Mobility Analyzer | TSI | 3085 |
| Dilution Bridge | Custom made | |
| DownToTen Sampling System | Custom made | |
| Electrostatic Classifier | TSI | 3082 |
| Hand held Mass Flow Meter (MFM) | Vögtlin | |
| miniCast Soot Generator | Jing Ltd | |
| Mobile Battery LPS 1500 | Clayton Power | |
| Nitrogen Gas Bottle | | |
| Propane Gas Bottle | | |
| Soft X-Ray Neutralizer | TSI | 3088 |
| Synthetic Air Bottle 10 L | | |

Comments/Description

Particle counter with a cut point of 4 nm

Mass flow controllers for controlling the miniCast gas flows

Device for the measurement of the exhaust flow rate of vehicles

Device for the removal of volatile compounds in an aerosol by oxidation

Oxidation and dilution air supply for miniCast

Particle counter with a cut point of 10 nm

Particle counter with a cut point of 23 nm

Part of the electrostatic classifier where the particle are separated by mobility.

Needle valve in parallel to HEPA filters. Used to adjust particle concentrations for calibration purposes

Custom made sampling system for the assessment of automotive sub-23 nm particle emissions

Device for the classification of aerosol particles by electrical mobility diameter

Device for measuring the inlet flow of measurement instruments

Combustion aerosol standard, soot generator

Battery for power supply of the DTT measurement system

Nitrogen for Mixing gas and quench gas supply of miniCast

Fuel for miniCast

Device for the establishment of the equilibrium charge distribution of aerosol particles

Gas Bottle for the dilution air supply

Editorial Comments

General:

1. Please take this opportunity to thoroughly proofread the manuscript to ensure that there are no spelling or grammar issues.

The manuscript has been thoroughly proofread.

2. Please ensure that the manuscript is formatted according to JoVE guidelines—letter (8.5” x 11”) page size, 1-inch margins, 12 pt Calibri font throughout, all text aligned to the left margin, single spacing within paragraphs, and spaces between all paragraphs and protocol steps/substeps.

It has been verified that the manuscript is formatted according to the JoVE guidelines.

3. Please avoid the use of equation editor for inline text, as such text will be formatted differently in the final manuscript.

This has been discussed with the editor.

4. Please rewrite lines 30-38 to avoid textual overlap with previous work.

The lines have been rewritten.

5. Please include at least 6 key words or phrases.

Keywords have been added.

Protocol:

1. The Protocol should be made up almost entirely of discrete steps without large paragraphs of text. Please move introductory paragraphs to the Introduction, Results, or Discussion, as appropriate.

The description of the measurement system was transferred to a separate section (Instrumentation). If the addition of this section is found to be inappropriate for this journal, the description of the system can be moved to the introduction.

2. For each protocol step/substep, please ensure you answer the “how” question, i.e., how is the step performed? Alternatively, add references to published material specifying how to perform the protocol action. If revisions cause a step to have more than 2-3 actions and 4 sentences per step, please split into separate steps or substeps.

Specific Protocol steps:

1. 1: Please include instructions in the imperative in this section.

The description of the measurement system was transferred to a separate section.

2. 3.3.3: Please include more information about your specific EFM installation if this is to be filmed.

We updated the manuscript and explained the EFM installation in more detail.

3. 3.3.8: What is your warm-up time?

The warm- up time of our EFM up to 15 min, depends on the ambient temperature

4. 5.2: How exactly is the alignment done? Is it just shifting the particle number graph by the transport time?

We changed the section of the time alignment. Normally we also record OBD data during our measurements, if possible, and shift the measurement data towards positive engine power. Since we only have the exhaust gas mass flow and the particle concentration as measured parameters, the particle concentration can also be shifted towards the exhaust gas mass flow. However, in this case one makes a more or less small mistake in the data evaluation, because as also noted by reviewer 3, high particle concentration does not only occur with high exhaust gas mass flows. A particle concentration peak usually occurs at the beginning of the acceleration phase, i.e. when the exhaust gas mass flow increases. Therefore, the revised manuscript describes a different method of time alignment. The particle concentration is simply shifted by the transport time from sampling point to analyser. This transport time has been calculated and explained in the new manuscript.

Figures:

1. Figures 11 and 12 are not cited correctly.

This has been corrected.

Table of Materials:

1. Please ensure the Table of Materials has information on all materials and equipment used, especially those mentioned in the Protocol.

Missing Items have been added.

Reviewer 1

Manuscript Summary:

The paper presents a portable sampling system accordingly designed to be used for PN measurements down to at least 10 nm. Moreover, it describes the system's installation and operation as a PEMS for RDE measurements. The paper is well structured and written. However, some parts of the manuscript require revision.

Major Concerns:

Introduction

1) Page 3, lines 115-117 & Page 4, lines 117-119: The authors mention that a lab-based system was first developed and then optimized for RDE measurements. The optimization of the PEMS version is described in 2 different parts of the introduction but could be described together. More importantly what misses is a more precise description of the actions that were taken in order to handle RDE measurements difficulties. For example the authors mention that the PEMS system is more robust for harsher vibration environments but no relative equipment is described in the M/S (CPCs are not influenced by vibrations?).

The two separate paragraphs have been unified. A short description of the pressure and acceleration robustness and references to according publications have been included.

Protocol

Description of the DTT PN-PEMS

2) General comment: Some important information miss in this section; the size of the DTT system (does it fit in small LDVs?-which are the trunk size requirements?) and the weight (it is referred in the discussion but would be more appropriate to be also mentioned in the description).

The according information has been added to the description of the system

3) page 5, line 188 (1.2 Catalytic stripper): The catalytic stripper is commercial? If not, is there any reference for its performance in removing tetracontane particles as required by the PMP protocol? Considering that C40 removal tests are not described in the calibration of the system, this is an important information that misses.

The information that the catalytic stripper is commercial has been added. We fully agree the the volatile particle removal efficiency should be explicitly mentioned. We performed tests using emery oil particles A short section about the volatile particle removal tests and the results has been added:

"The volatile particle removal efficiency of the CS was verified with polydisperse emery oil particles of > 50 nm and > 1 mg/m³ (3.5 to 5.5 mg/m³) showing efficiency of better than 99% (actual value 99.9%) as defined in RDE regulation (2017/1154). This is a more severe test than the tetracontane test prescribed in the current PMP protocol."

4) page 5, line 215 & 217 (1.8 & 1.9): How long can you measure with this battery and this synthetic air bottle?

The according information has been added.

Calibration procedure

5) page 8, line 343: The authors calculate the average penetration fraction and subsequently the average PCRf according to the current PMP protocol. Their rationale is understandable (comparison with commercial PN23 measurement systems). However, the authors should (at least) mention somewhere in the M/S which is the d50 of the sampling system. This information is shown in Figure 11 but is not clear.

The d50 has been added in the discussion and the is explicitly marked in the figure.

Installation and preparation for real driving emissions measurements

6) page 10, line 388: There is no discussion about the safety in the vehicle cabin due to the working fluid of the CPCs.

A description of how the butanol bottles have to be mounted is added in the manuscript.

7) page 10, line 392: "recalibrated AVL CPC" Which model? How was it recalibrated? Please mention briefly the method (T saturator/condenser change?).

The information that this CPC was recalibrated was not correct. The CPC used was a commercially available 10 nm AVL CPC. This has been corrected accordingly

8) page 10, line 419: The CPC is connected to an external vacuum pump. The outlet of the pump is driven outside the vehicle?

Yes, the outlet of the pump is driven to the outside of the vehicle. This is now explicitly mentioned in the manuscript.

Minor Concerns:

Introduction

1) Page 3, lines 100-102: It would be beneficial to refer the relative legislation: EURO 5b for diesel, EURO 6 for G-DI etc.

We fully agree. The respective standards have been added.

2) Page 3, line 105: Reference 10 misses publication year and volume

The reference entry has been corrected

3) Page 3, line 121-122 (Table 2): It is not clear what the table compares in stage 0. Stationary (PMP) vs PEMS (DTT)? Moreover, it is not clear the stage 1 (particle transport): "to be specified in DTT".

We agree that this table is partly out of context and does not contribute to the readers understanding. Therefore, Table 2 has been removed.

Protocol

Description of the DTT PN-PEMS

4) page 4, line 172 (Figure 2): This figure does not show a picture of the measurement system but only of the sampling system. Please correct the legend and the relative text. Moreover, the various parts of the system could be indicated with legends inside the figure.

The legend has been corrected. The figure has been a modified to indicate the parts with legends.

5) page 4, line 175 (1.1 Dilution): Which is the total DR of the system? The hot dilution T is mentioned later in the M/S but it would be useful to report it also here.

We agree that it is useful to mention both the DR and the hot dilution T here. Typical values for both paramters are mentioned now. Furthermore, a formula how to calculate the dilution ratio is provided.

6) page 5, line 178: "Low particle loss characteristic"-Reference?

References have been added.

Calibration procedure

7) page 7, line 272 (Table 3): The legend should be corrected: Suggested instead of suggest. Moreover, in Table 3 the nitrogen flow is the mixing gas flow?

The legend has been corrected. It has been clarified that the nitrogen flow is the mixing gas flow :
Mixing gas (N₂)

8) page 9, line 347: DR instead of D

This has been corrected.

Measurement operation

9) page 11, line 451: Differ instead of diluter

This has been corrected.

Data analysis

10) page 12, line 504 (Figure 10): There is a line over the figure.

We cannot confirm this for our version of the submitted manuscript. We will make the editor aware that you experienced an issue with this figure and ask if there is anything we can do to avoid this.

Representative results

11) page 14, lines 582-583: The discussion is too simplistic for a so wide and complex topic. I would suggest either to avoid the discussion or to cite at least review papers on this topic (for example Giechaskiel et al. 2014).

We agreed that the discussion of this topic is beyond the scope of this article. This has been explicitly stated and references have been provided for further information.

Discussion

12) page 15, line 633: Apart from the EFM did the authors use also other instrumentation for the exhaust mass flow rate measurement?

It was intended initially to also include a different option for the determination of the exhaust mass flow but we decided to keep the focus on the EFM to keep the paper more concise. This sentence was removed now.

13) page 16, line 656: Particle losses are low compared to what? The low loss characteristic should be discussed in terms of d₅₀.

We agree that it is better to mention the d₅₀ here instead. The sentence has been changed.

Reviewer 2

Manuscript Summary:

The paper addresses the timely topic of tailpipe particle emission measurement methodology on a car. Particularly in Europe, there is an increasing interest in particle number (PN) measurement, as discussed in sub-23 nm PN measurement by the Particle Measurement Program (PMP) in the Working Party on Pollution and Energy (GRPE), under the auspices of the United Nations Economic Commission for Europe (UN ECE). This paper is discussing about sub-23 nm PN measurement according to using portable measurement techniques with several vehicle testing.

Major Concerns:

The variable and limited experiments provide reliable results and a fundamental concept for the characterization of sub-23 nm PN measurement. This paper is well-structured and clear. It is helpful and acceptable for the audience of Journal of Visualized Experiments, but not without substantial revision. I will recommend revising some descriptions to maintain of the quality of manuscript.

We revised the manuscript substantially and clarified wordings and descriptions.

Minor Concerns:

P17 of 18, Line 703: (0) 0, 1-16 -> 51 (5), 626-641.

The reference entry has been corrected.

Reviewer 3

Manuscript Summary:

Interesting manuscript describing the apparatus for measurement of very small particles in an internal combustion engine exhaust. Reasonably well written, comprehensive, with good introduction.

Major Concerns:

No specific issue, but a large number of minor problems. Additional read-through by the authors recommended. References need additional attention so that the work can be readily located.

Minor Concerns:

Define all abbreviations - DPF on line 60, DETR on line 65, and throughout the work
Missing definitions have been added.

Line 78 - while respecting the reference to which the 20 nm size is attributed, the primary particle size can be much lower, around 10 nm, i.e. Liati et al. around 2010, Ronkko et al. EST around 2013, presentations at ETH Conference on Combustion Generated Nanoparticles, ...

We agree that the primary particle size can be much lower. The wording of the paragraph has been changed to include this information and the according references.

Line 87 - definition of a regulatory particle - there is no representative particle, only boundary conditions on particle size and volatility

We agree that this should be described more precisely. The wording has been changed accordingly.

Fig. 2 and other figures with photos - arrows pointing to specific parts and some legend would be helpful

We agree that this is very helpful. Legends and arrows have been added.

line 200 - Mass flow management - two flows are controlled, two are only measured - in my opinion a total of three of the four flows should be controlled for a two-stage dilution using porous tube diluters.

It is correct, that with the mass flow control system, it is not possible to control each flow separately. As described in the manuscript, the difference between the dilution air flow and the respective excess flow can be defined in the software and the absolute level of the flows is controlled manually with needle valves downstream the mass flow meters. To make this more clear, the needle valves have been added in figure 3. With this arrangement the dilution ratio is not perfectly constant over time, but drifts are corrected for in the post processing. This arrangement was chosen because the mass flow controllers used exhibit a relatively high pressure drop, which would have required the use of a more powerful pump. As a result, this would have increased the power consumption of the overall system significantly.

line 340 - a note that each counter works with a different dilution ratio, and specifying what dilution ratio (that in a dilution bridge, not dilution ratios of porous tube diluters?) would be helpful

We agree that it is helpful to clarify that this is the DR from the porous tube diluters. The DR from dilution bridge is not relevant because it applies for both CPCs. An according sentence has been added.

line 495 - How are you sure that high exhaust flow correlates to high particle concentrations? What if you have peak concentrations at the beginning of the acceleration, before the flow peaks? We changed the section of the time alignment. Normally we also record OBD data during our measurements, if possible, and shift the measurement data towards positive engine power. Since we only have the exhaust gas mass flow and the particle concentration as measured parameters, the particle concentration can also be shifted towards the exhaust gas mass flow. However, in this case one makes a more or less small mistake in the data evaluation. Therefore, the revised manuscript describes a different method of time alignment. The particle concentration is simply shifted by the transport time from sampling point to analyser. This transport time has been calculated and explained in the new manuscript.

line 530 - Fig. 11 and Fig. 12 are swapped
This has been corrected

line 605 - legends are mixed up - no legend for Fig. 10, what is given for Fig. 10 should be for Fig. 11, for Fig. 11 should be Fig. 12.
This has been corrected

line 633 - two different methods - only one is described - what is the other one?
It was intended initially to also include a different option for the determination of the exhaust mass flow but we decided to keep the focus on the EFM to keep the paper more concise. This sentence was removed.

line 675 - check author name
The reference entry has been corrected.

references - 6, 8, 10, 11, 15, 18-20 - unify citations of EU legislation, when referring to www sites please specify the exact location in the URL link, provide enough information so that the reader can easily find the work, ref. 10 bad volume, ref 8 unclear title, ref 24 refer to the official conference proceedings at nanoparticles.ch
The reference entries have been corrected/modified accordingly

Reviewer 4

Manuscript Summary:

This paper reports on the experimental implementation of a research tool called DownToTen for wide-size-range particle measurements. The tool is mobile and can thus be used e.g. for in situ on-road vehicle particle emission measurements among many other applications. The detailed set-up of the instrument is demonstrated. In addition, some critical alignment and calibration parameters and procedures are discussed.

Major Concerns: None

Minor Concerns:

The paper is nicely and clearly written. The authors are highly qualified and respected in the community; this paper adds to this notion.

I am not familiar with this publication format though. The review invitation letter pointed to some sort of video format, which also explains the content organization. As such, the paper should be accepted with only minor changes to the wording.

There is one single comment on the summary. I believe that addressing the adverse health effects of ultra-fine particles at this point is not necessary, particularly when there is no reference provided. This discussion is currently entirely open; the authors phrase their concern accordingly: "Studies suggest that sub-23 nm particles might be even more harmful to humans than larger particles". This sentence should either be backed by references or deleted. In the introduction on the other hand, the reasoning by the authors is nicely put into perspective.

We agree that this sentence is not necessary at this point. The sentence has been deleted.