## EMRP ENV02 PartEmission

WP2 Task 2.2 Deliverable 2.2.2

User's handling experience with the instrument selected in deliverable 2.2.1 and testing results to analyse the instrument's long term stability and operational reliability

Final: 2014-07-14

Anke Jordan-Gerkens, Andreas Nowak, Arne Kuntze, Margit Hildebrandt (PTB)<sup>1</sup> Francesco Riccobono (JRC)<sup>2</sup>

<sup>&</sup>lt;sup>1</sup> Physikalisch Technische Bundesanstalt (PTB), Germany <sup>2</sup> Joint Research Centre (JRC), EU

# **CONTENT**

1	Introduction	3
2	User's handling experience	4
2.1	Measurements under service conditions	4
2.2	Measurement setup at DEKRA	4
2.3	Results	5
3	Long term stability and operational reliability	8
3.1	Measurement setup	8
3.2	Measurement procedure	9
3.3	Results	10
4	Conclusions	12
5	ANNEX A	14
6	References	17

### 1 Introduction

Vehicles are a major source of particulate matter (PM) in urban areas causing adverse health effects. The European legislation regulates the PM emissions from light duty vehicles with the UNECE Regulation No. 83 setting limits for the emitted mass and number of particles per kilometer. With the entry in validity of the particle number (PN) limit for compress injection (diesel) light duty vehicles (September 2011) the diesel vehicles are equipped with Diesel Particulate Filters (DPF) which assure the compliance with the new PN limit and led considerable reduced particulate emissions from modern diesel vehicles.

The regulated measurement principles for type approval test in Europe are the gravimetric collection of particles of filter (for PM) and the light scattering measurement of the single particles after growth of the particle size to detectable sizes by means of a Condensation Particle Counter (CPC). In addition, in order to measure only the non-volatile fraction of the particles, a pre-conditioning unit is coupled with the CPC, which removes the volatile fraction through a hot dilution and a Volatile Particle Remover, a section of tube kept at least 300°C. Currently the regulation requires that the periodic emission control of the emission of diesel vehicles is performed with an opacimeter. The opacimeter is an instrument based on the measurement principle of light extinction; however modern diesel vehicles (equipped with DPFs) emit a concentration of particles which is often close to the detection limit of instruments based on light extinction.

The aim of the EMRP ENV02 "PartEmission" Work package 2 "Evaluation of measuring methods for particle emission from modern diesel vehicles in periodic emissions control" is to develop a metrological background for validation of novel instruments measuring the concentration of combustion particles in exhaust gases from diesel vehicles, which can be used for the regulatory periodic emissions control of vehicles. In Task 2.1, the available prototypes and potential future measurement principles were summarized and aligned with specific requirements concerning periodic emission control [1,2]. Task 2.2 is the trial of the suitable instruments, determined in task 2.1, in field tests and their comparison with the current standards.

This report summarizes the actions of the project partners investigating the practical usability tests and long term drift due to soot deposition with capable instruments. The results of this report fulfill the deliverable 2.2.2 from ENV02 "PartEmission".

#### 2 USER'S HANDLING EXPERIENCE

The evaluation of the user handling experience of the instruments under test in the frame of the ENV02 WP2 was carried out at DEKRA in Stuttgart, Germany. For this tests five instruments, which were evaluated in the previous steps of the project [1,2], were at our disposal.

#### 2.1 Measurements under service conditions

Three instruments based on the light scattering principle (L) determine particle mass as well as opacity by passing the particles through a laser beam and measuring the scattered light at different angles. These instruments are designed for the measurement of exhaust emissions with particle sizes above 80 nm and are more sensitive by a factor of 100 than the established opacimeters.

The instrument based on diffusion charger principle (DC) determines the particle mass, number and surface concentrations. These values can be determined by charging the particles and subsequently collecting them and measuring the total current deposited on the filter. This instrument, equipped with a dilution unit, was the only instrument capable of measuring particles below 100 nm reliably at representative particle mass and number concentrations of modern diesel emissions according the Swiss regulation (SR 941.242) in the previous laboratory tests. A second DC instrument couldn't be evaluated because of a malfunction of the instrument<sup>3</sup>.

The ionization chamber (IC), a non-commercial instrument, is measuring total particle length using a set of commercial smoke detectors. The conversion from total length to particle number [3] was made by assuming a mean particle diameter of 80 nm and a particle size distribution with geometric standard deviation (GSD) of 1.8. Moreover, the zero calibration had to be performed after the measurements, as the automated calibration was not yet included in the instrument software available at the time of the tests.

The measurements were performed as usual periodic emission tests at DEKRA with three different vehicles. The first was a low emitting Audi A4 (Euro 5), the second a VW Passat (Euro 6) with a broken DPF and the third a VW Multivan (Euro 4) with an upgraded DPF (certificates of registration are reported in Figures A2, A3 and A4 respectively).

## 2.2 Measurement setup at DEKRA

The sampling was realized simultaneously for all instruments using an extension tube connected to the exhaust pipe of the vehicles on which the single sampling tubes of the instruments were clamped on (Figure 1). All sampling tubes of the instruments were connected at the same time. The total sampling volume of the instruments was 67 l/min ca. (L1: 2.4 l/min, L2: not measurable, excess pressure, L3: 2.6 l/min, DC1: 4.2 l/min, IC: ca. 15 l/min, Reference Opacimeter: 42 l/min). It is assumed that a sufficient sampling was

<sup>&</sup>lt;sup>3</sup> After the laboratory tests the manufacturer found a non-soot contamination of the instrument. A replacement of the sensor for these tests wasn't possible because of time constraints.

guaranteed by the ratio sampling volume to exhaust volume, since the exhaust flow was always larger than the sampling flow. Problems which could occur through an insufficient dynamic pressure during the sampling can be excluded, because the only instrument which works according this principle showed the best results when compared to the reference instrument.

After warm up, the accelerator pedal was fully pushed 5 times. This procedure was repeated 3 or 5 times depending on the test vehicle, leading to 3 or 5 mean values of the peak absorption coefficient and the biggest difference between the single values measured by the highly sensitive reference opacimeter (AVL 439) at DEKRA<sup>4</sup>. Here the measuring mode of the opacimeter was changed between mode 1 (Peak value, mode A, without filter) and mode 2 (ECE R24, EEC 72/306) which is commonly used in usual periodic emission tests. Note that the measuring mode 2 leads to significantly lower absorption coefficients compared to mode 1 in the response of the reference opacimeter because of the different ranges for the calculation of the mean value [4].



Figure 1: Sampling setup during the practical usability tests at DEKRA; single sampling tubes for each instrument were clamped on the extension tube connected to the tailpipe

#### 2.3 Results

The first test vehicle was a low emitting Audi A4 (Euro 5). During the three test cycles the reference opacimeter measured very low absorption coefficients from 0,003 m<sup>-1</sup> (mode 1) to 0,001 m<sup>-1</sup> (mode 2). The light scattering instruments had no response for the three acceleration cycles. There was some response from the diffusion charger but it couldn't be

<sup>&</sup>lt;sup>4</sup> The Calibration of the instrument was performed with traceable gray filters from PTB (see annex, table A1)

related to the acceleration cycles. Only the ionization chamber has shown a response of  $1.6 \cdot 10^5$  and  $2.3 \cdot 10^5$  particle/cm³ for the first two acceleration cycles corresponding to an absorption coefficient of  $0.003 \text{ m}^{-1}$  (Figure 2). It is expected, that the opacimeter response (very low absorption coefficients) is due to the cross sensitivity of gaseous components in the exhaust while the IC response is due to volatile components because this instrument was not equipped with a heated sampling line.

The second test vehicle was a VW Passat with a broken DPF with a comparatively high particle concentration in the exhaust (Figure 3). The reference opacimeter showed absorption coefficients of approximately 1.4 m<sup>-1</sup> for the first three acceleration cycles (mode 1). The switch to the mode 2 led to a decreasing response of the opacimeter to a value comprised between 0.76 m<sup>-1</sup> and 0.84 m<sup>-1</sup>.

The three light scattering instruments showed different response. L1 displayed a relatively low adsorption coefficient between 0.25 and 0.3 m<sup>-1</sup>. The indicated values of instrument L2 (0.65 up to 0.85 m<sup>-1</sup>) matched well with the reference values of measuring mode 2. The displayed absorption coefficients of L3 were relatively low and showed a higher variability than the other light scattering instruments, ranging from 0.22 to 0.47 m<sup>-1</sup>.

The particle number concentration measured by DC1 was  $(1.95 \cdot 10^8 \text{ to } 2.3 \cdot 10^8 \text{ #/cm}^3)$  more than one order of magnitude higher than the concentration measured by the IC  $(0.96 \cdot 10^7 \text{ to } 1.07 \cdot 10^7 \text{ #/cm}^3)$ . Because there was no particle number reference during this test, there was no information about the actually particle number concentration in the exhaust.

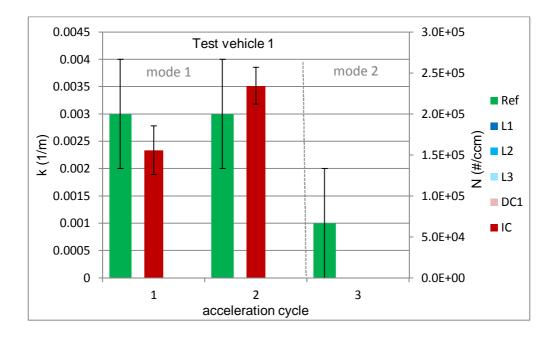


Figure 2: Response of the reference instrument (opacimeter, AVL 439. DEKRA) and the measuring instruments under test (L-Light scattering instruments, DC-Diffusion Charger, IC- Ionisation Chamber) to the emitted particle concentration (primary vertical axis: absorption coefficient in m<sup>-1</sup>, secondary vertical axis: particle number concentration in particle/cm<sup>3</sup>) of the test vehicle 1 (Audi A4, Euro 5)

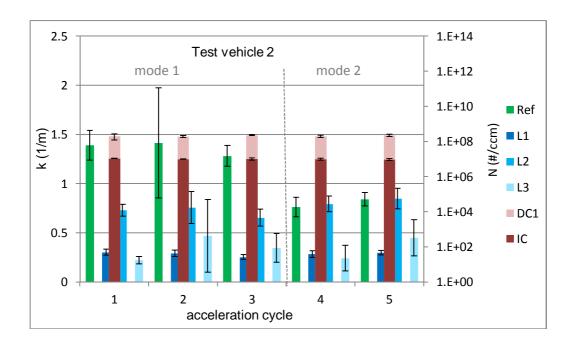


Figure 3: Response of the reference instrument (opacimeter, AVL 439. DEKRA) and the measuring instruments under test (L-Light scattering instruments, DC-Diffusion Charger, IC- Ionisation Chamber) to the emitted particle concentration (primary vertical axis: absorption coefficient in m<sup>-1</sup>, secondary vertical axis: particle number concentration in particle/cm<sup>3</sup>) of the test vehicle 2 (VW Passat, Euro 6, with broken DPF)

A VW Multivan, Euro 4, with upgraded DPF was the third test vehicle. As shown in Figure 4 the reference opacimeter showed absorption coefficients of  $0.86~\text{m}^{-1}$  (mode 1) for the first acceleration cycle. The switch to the mode 2 led to a decrease of the response of the opacimeter to  $0.37~\text{m}^{-1}$  and  $0.33~\text{m}^{-1}$ .

In this particle concentration range the instruments under test showed a similar response to the opacimeter and also the values measured by the different instruments under test are more consistent to each other compared to the tests performed with the two previous vehicles.

L1 displayed relatively low adsorption coefficients comprised between 0.42 and 0.43 m<sup>-1</sup>. In the last two cycles (mode2), instrument L2 (0.37 up to 0.65 m<sup>-1</sup>) measured adsorption coefficients which resulted to be the closest to the values measured by the reference instrument. The displayed absorption coefficients of L3 are relatively low and show a larger variability than the variability of the other light scattering instruments, ranging from 0.16 to 0.27 m<sup>-1</sup>.

The particle number concentration measured by DC1 was, similarly to what observed before,  $(1.1\cdot10^8 \text{ to } 1.2\cdot10^8 \text{ #/cm}^3)$  about one order of magnitude higher than the concentration measured by the IC  $(1.2\cdot10^7 \text{ to } 1.5\cdot10^7 \text{ #/cm}^3)$ . Since there was no particle number reference instrument during this test, there was no information about the actual particle number concentration emitted by the vehicles.

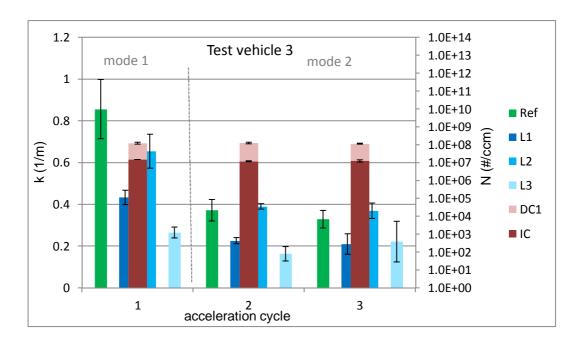


Figure 4: Response of the reference instrument (opacimeter, AVL 439. DEKRA) and the measuring instruments under test (L-Light scattering instruments, DC-Diffusion Charger, IC- Ionisation Chamber) to the emitted particle concentration (primary vertical axis: absorption coefficient in m<sup>-1</sup>, secondary vertical axis: particle number concentration in particle/cm<sup>3</sup>) of the test vehicle 3 (VW Multivan, Euro 4, with upgraded DPF)

#### 3 Long term stability and operational reliability

## 3.1 Measurement setup

The PTB-setup for the evaluation of instruments in laboratory tests included a modified HiMass-CAST (L. Jing, Zollikofen, Switzerland) as soot generator, a special developed dilution system for the addition of particle free dilution air under controlled conditions of humidity and temperature, a conditioning unit and a sample splitter with 10 sample ports for the reference and monitoring instruments as well as devices under test (see Figure 5 for details on the setup).

The test aerosol was generated in 19 diffusion flames with controlled propane to air ratio. Then the soot formation was quenched with a controlled nitrogen flow. This allowed a controlled adjustment of the generated particle size and size distribution. The soot generator output flow rate of 450 l/min was diluted in a self-developed diluting system (counter flow mixer). This ensured proper mixing of the aerosol and the particle free dilution air, under controlled humidity and temperature conditions, with an adjustable ratio. The absolute aerosol flow after the dilution unit was around 900 l/min depending on the set dilution ratio. In the following conditioning unit the soot aerosol was aged and stabilized. Using this soot generator configuration a soot aerosol with GMD 50-240 nm and GSD 1.7-2.2 was provided for the instrument evaluation.

The sample flow splitter allows sampling of 10 different instruments at 10 sample ports at the same time. In order to decrease the input concentration a rotation disc thermodiluter with a

dilution factor 1:1000 was located upstream of a SMPS-System and a CPC (marked as EECPC in

Figure 5). The SMPS system was used for measuring the particle number size distribution of the soot aerosol and the CPC was used to measure the number concentration. The gravimetrical mass of the soot aerosol was determined by filter sampling. As reference a laboratory instrument measuring the opacity (Opacimeter AVL 439, Graz, Austria, fully characterized at PTB) was used. The sample flows of the instruments were the following: Reference opacimeter AVL 439 16 l/min, rotation disc and thermodiluter approximately 2 l/min (Dilution 1:1000) [4].

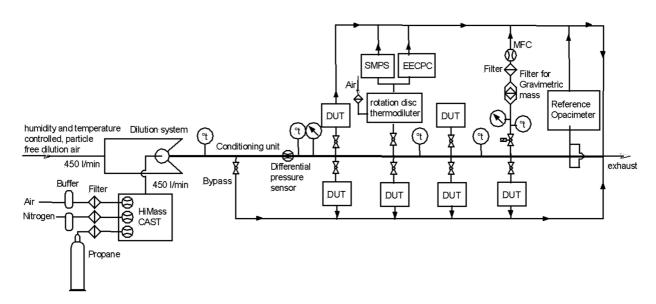


Figure 5: Experimental setup used for evaluating performance of particle measuring instruments at PTB

## 3.2 Measurement procedure

The instruments were evaluated with a soot aerosol with an opacity between 0.01 and 1.2 m<sup>-1</sup>, particle sizes between 50 and 237 nm and particle number concentrations ranging from 1.16·10<sup>7</sup> to 1.1·10<sup>8</sup> cm<sup>-3</sup>. This corresponds to a particle mass concentration of around 3 mg/m<sup>3</sup> to 380 mg/m<sup>3</sup>, which is much higher than the known emission ranges of modern diesel vehicles. In order to cover the wide opacity range of 0.01 to 2.98 m<sup>-1</sup> particle size (and such particle mass concentration) and particle number concentration were varied in parallel. The aerosol characteristics were adjusted by changing the propane-, air- and nitrogen flow within the soot generator. Note, although the opacity of modern diesel vehicles is commonly much lower than 1 m<sup>-1</sup> this wide measurement range was chosen by PTB as this is the current required range for an opacimeter.

For the light scattering instruments efficiency is defined as the interrelation between the opacity displayed by the instrument and the opacity of the reference opacimeter of PTB.

The correlation between the measured opacity by the devices under test and the measured opacity by the reference opacimeter of PTB was determined [2].

#### 3.3 Results

In order to determine the long term stability and operational reliability of the instruments, the results which were presented in the report Deliverable 2.1.2 of this WP were compared with the results of the measurements which were performed one year later.

Figure 6 shows the results of the light scattering instruments and the diffusion charger instrument DC1 against the reference opacity of PTB in the range from 0.015 up to 1.2 m<sup>-1</sup>. The measurements were performed at PTB in February 2013 and repeated in February 2014 after the laboratory tests took place at PTB, METAS and MIKES and after the field tests at JRC. IC and DC2 were not available during the measurement campaign of 2014 so there is no direct comparison with the measurement campaign of 2013.

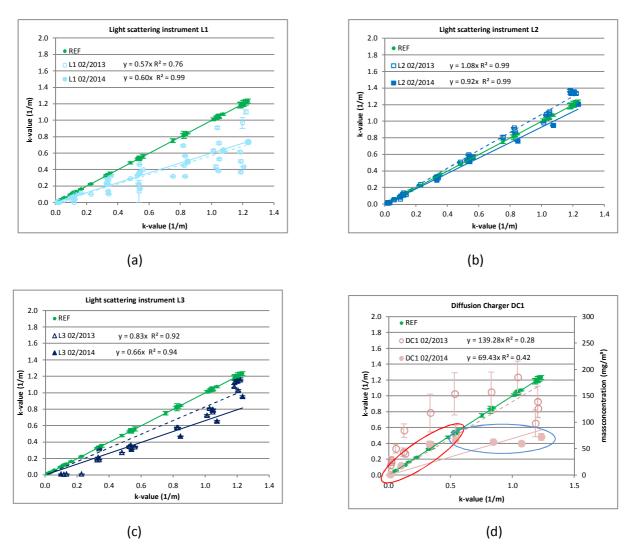


Figure 6: Comparison of the results from the opacity measurements at PTB in 02/2013 and 02/2014 The results of the light scattering instruments L1 (a), L2 (b), L3 (c) are plotted against the left y-axis including a linear fit (y=mx) and coefficient of determination. The mass concentration of the diffusion charger, DC (d) is plotted against the right y axis. For the devices under test, the error bars denote the standard deviation.

The light scattering instrument that showed the best correlation with the reference value of absorption coefficient after one year is L2, which confirmed the high coefficient of determination (0.99) measured in 2013 and showed a decrease of the slope m from 1.08 to 0.92 (Figures 6b). L1 improved significantly the linearity showing a much lower dispersion of the data after one year (the coefficient of determination changed from 0.76 to 0.99) although the slope changed only slightly from 0.57 to 0.60, still far from the reference value (Figures 6a). The light scattering instrument L3 showed a slight improvement of the dispersion (the coefficient of determination changed from 0.92 to 0.94) but the slope changed by -20% reducing the agreement with the reference value (Figure 6c). The slope calculated in 2014 for the diffusion charger instrument DC1 showed a much larger deviation compared to the 2013 values. One possible reason for this larger deviation is the fact that the sensor of the diffusion charger was replaced by the instrument manufacturer between the two measurements campaigns due to a failure of the sensor itself. Moreover the disagreement between the DC1 and the reference (coefficient of determination changed from 0.28 to 0.42 in 2014) is possibly due to the fact that the diffusion charging instruments are built to measure particle number (or particle surface) concentration and not opacity, especially at relatively high opacity values (> 0.5 m<sup>-1</sup> high particle concentration) the DC reached saturation (as highlighted by the blue ellipse in Figure 6d). Excluding the values of opacity > 0.5 m<sup>-1</sup> the correlation with the reference instrument for 2014 improved compared to the correlation assessed in 2013 (as highlighted by the red ellipse in Figure 6d).

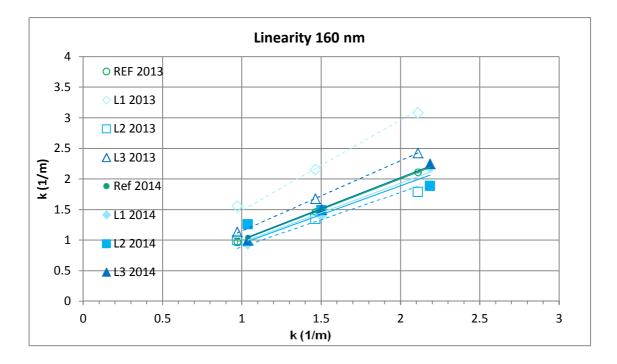


Figure 7: Linearity of candidate instruments responding to 160 nm soot particles. Comparison of the results from the opacity measurements at PTB in 02/2013 and 02/2014.

Figure 7 shows the linearity for opacity that was tested at PTB using CAST aerosol at 160 nm aerosol size. The opacity values used for this linearity test are much higher than the ones that can be expected for modern diesel vehicles. However the comparison of the data

collected in the measurement campaigns of 2013 and 2014 shows that the response of the instruments was not the same after the measurements at METAS, MIKES and JRC. In Table 1 are reported the coefficient m of the linear fits applied to the data of Figure 7, and the corresponding coefficients of determination. The slopes m of L1, L2 and L3 changed by -35%, 5% and 12% respectively. All the instruments improved the correlation with the reference instrument (see changes of slopes m in Table 1 from 2013 to 2014). It is particularly relevant the improvement of L1 that significantly reduced the offset from the reference values (Figure 7).

Table 1: Fitting value m and  $R^2$  corresponding to the linear fits of Figure 7 for L1, L2 and L3 for the measurements performed in 2013 and 2014.

	2013		201	4
	m	$R^2$	m	$R^2$
L1	1.48	0.98	0.96	0.98
L2	0.89	0.91	0.94	0.44
L3	1.15	0.99	1.01	0.99

### 4 CONCLUSIONS

All instruments resulted transportable, easy to use and all were able to sample the vehicles exhaust with simple installation at tailpipe. The results of the tests with the low emitting Euro 5 vehicle (equipped with a fully functional DPF system) has shown that the emitted particle concentration was below the detection limit of the instruments. The difference between the results of the diffusion charger (DC1) and the ionisation chamber (IC) for the test vehicle 1 could be explained by the presence of the volatile particle remover (VPR) in the DC1 that removed the most volatile particles. Despite the presence of the VPR the particle emissions of the other two test vehicles measured by the DC1 was more than one order of magnitude higher than what measured by the IC. Due to the missing particle number reference during this test, there was no information about the actually particle number concentration in the exhaust so that it cannot be stated which instrument shows a realistic response, the accuracy of the particle number instruments is addressed in Deliverable 2.1.2 where the instruments for periodic inspections measuring the particle number are compared to a reference particle number counter. The results at DEKRA showed also that instruments working with the light scattering principle are able to detect malfunctions of the exhaust after treatment system of the Euro 6 vehicle and measuring particle concentrations of the Euro 4 vehicle.

The correlation of the light scattering instruments and the diffusion charge instrument DC1 with the reference instrument was calculated from data collected in 2013 and in 2014. The large changes observed from 2013 to 2014 in the linear correlations of some of the light scattering devices indicate that the recalibration of the light scattering instruments in needed more frequently than once a year. Additional measurements should be performed at shorter

time distance in order to determine the exact maximum period for the recalibration of the light scattering instruments. The long term stability of the diffusion charge instrument DC1 was assessed against the reference opacimeter, showing an improvement of the performance of the diffusion charge instrument in 2014 compared to 2013 (probably also due to the replacement of the internal sensor). The correlation improved remarkably especially for values of absorption coefficient smaller than 0.5 m<sup>-1</sup>, above this value the DC1 instrument reached saturation. The instruments based on diffusion charging to assess emissions from vehicles are currently undergoing a great boost in their quality also due to the EC program aimed to measure particulate number emissions with on-board measurement systems. Additional check of their long term stability should be repeated in the near future to assess the performance of the more advance prototypes and series products.

# 5 ANNEX A

Table A 1: Calibration of the reference opacimeter AVL 439 (DEKRA) with gray filters

FilterNr.	Sign of calibration	target N (%)	target k (1/m)	actual k (1/m)	u(k=2)
SN 1519	87099-PTB-09	10.5	0.257	0.2377	0.0024
SN 4460	87100-PTB-09	20	0.519	0.5267	0.0033
SN 4475	87101-PTB-09	39.7	1.176	1.2039	0.0017
SN 502086	87102-PTB-09	51.4	1.678	1.7304	0.0067

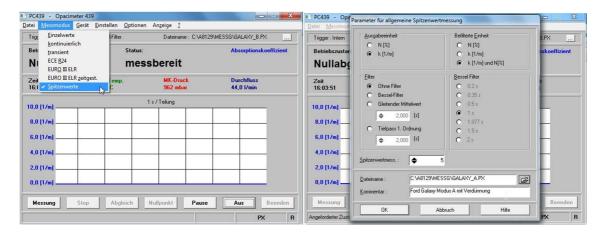


Figure A1: Settings of the reference opacimeter AVL439: measuring mode 1 – peak value without filter; measuring mode 2 – ECE R24

Europear	
Zulassungsbescheinigung Teil (	09.01.2014     0588     AHM000647     2     1     88/44

Figure A2: Certificate of registration of test vehicle 1 – Audi A4, Euro 5



Figure A3: Certificate of registration of test vehicle 2 – VW Passat, Euro 6 (broken DPF)

Zulassungsbescheinigung Teil I	21.11.2000 21 0603 22 45802D9	0111/04000 181	
(Fahrzeugschein)	21 0500	" 04789 " 1840	
S-0-165/13-00499	WV2ZZZ70Z1H081084 1 6	1900 6 02050	
Europäische D Bundesrepublik		u _ u _ o _	
Gemeinschaft Deutschland Permas de prodaeún Para I / Owedden o regulraci - Cant I /	7DZ	206 8 002810 0 002810	
Registro infrantesis. Del I / Registreer infistumistus. Dea I /		23 01510 23 01300 24 -	
Registrated certificate, Part I / Certificat d'immersociation, Partie I / Carta di predicatene, Parte I / Registragas apieciba, L divis /	-	01510 12 01300 12 -	
Registracijou laudijanus, i dats / Forgatni empedety t. Best / / Certifikas ta Registracijani, L-i Parti / Kentekenlowijs, Deel L / Object Rejistracijani, "Capit / Certificado de matricala Parta / /		w 78 w - w 74	
Coverticinie e sveningo. Cast 17 Prometria dovoljenja. Del 17 Relistoriërithedutus. Osa 17 Registraringsbeviset. Del 1	MULTIVAN	02000 02 0700 11 006 12 -	
A Ancika) is Feynolishum	VOLKSWAGEN-VW	150 205/60R16C100/98T	
-	SO.KFZ WOHNM.UEB.2,8 T	52 205/60R16C100/98T	
C.1.5 Naive of cr First enems.	GESCHLOSSEN	da	
The state of the s	9	[ ] N	
	98/69/EG III;A	x e1*98/14*0095*03	
C1.2 Vuoringia	DIESEL	6 01.08.2000 WB653266	
And the second s	0002 0654 02461	21 -	
C1.3 Andalli C	TO SECURE THE SECURE T		
70565 STUTTGART			
Novet and Jakes STUTTGART			
06.2015 ( bring 14.06.2013			
C.4c Der teneher der Zuläbungsbeschenigung wird eldet als Eigentümm des Fahrzeuts zurspreibnen.			
Tayravajs assignainses.	BR.II. ODBADANO ARAGONIO	DESCRIPTION OF PROPERTY	

Figure A4: Certificate of registration of test vehicle 3 - VW Multivan, Euro 4 (upgraded DPF)

#### 6 REFERENCES

- 1 Mamakos, et al., Report WP2 D1, Consistent requirements specified for novel measuring instruments (prototypes) and comparison with European legislative requirements, February 2012, http://www.ptb.de/emrp/partemission-publications.html
- 2 Spiegel, et al., Report WP2 D1.2, Laboratory evaluation of novel soot sensors for periodic emission control of modern diesel vehicles, April 2012, http://www.ptb.de/emrp/partemission-publications.html
- Litton, C. D., Smith, K. R., Edwards, R. & Allen, T.; Combined optical and ionization measurement techniques for inexpensive characterization of micrometer and submicrometer aerosols. Aerosol Science and Technology, 38(11), pp. 1054-1062, 2004
- 4 AVL List GmbH, AVL 439 Opacimeter, User manual AT2307D, Rev. 04, Graz, Austria, 2006