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VERTdePN – quality verification of combined DPF+SCR systems for retrofitting.

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ABSTRACT

New Diesel exhaust gas aftertreatment systems, with combined DPF *) and deNO$_x$ (mostly SCR) systems represent a very important step towards zero emission Diesel fleet.
These combined systems are declared today by the OEM's as an ultimate solution and are already offered by several suppliers for retrofitting.

Reliable quality standards for those quite complex systems are urgently needed to enable decisions of several authorities.

The Swiss Federal Office of Environment BAFU and the Swiss Federal Roads Office ASTRA decided to support further activities of VERT to develop appropriate testing procedures and to define the quality criteria for dePN systems.

The present report informs about the international network project VERT *) dePN (de-activation, de-contamination, disposal of particles and NO$_x$), which was started in Nov. 2006 with the objective to introduce the SCR-, or combined DPF+SCR-systems in the VERT verification procedure.

Examples of results for some of the investigated systems are given. These investigations included parameters, which are important for the VERT quality testing: besides the regulated gaseous emissions several unregulated components such as NH_3, NO_2 and N_2O were measured. The analysis of nanoparticle emissions was performed with SMPS and NanoMet.

The findings from the tested systems can be summarized as follows:

- the investigated combined dePN systems (DPF+SCR) for dynamic engine application efficiently reduce the target emissions with deNO$_x$-efficiencies up to 92% (if operated in the right temperature window) and particle number filtration efficiency up to 100%,
- the ammonia slip can be efficiently eliminated by the slip-cat,

*) Abbreviations see at the end of paper

- during the transient tests there are temporarily increased emission of NO and NH_3 due to momentary imbalance of the deNOx stoichiometry,
- in the configuration with urea dosing after DPF, a secondary formation of nanoparticles is detectable with a moderate increase of number concentrations but no critical impact on the overall filtration efficiency of the system,
- the average NO_x conversion rate at transient operation (ETC) strongly depends on the exhaust gas temperature profile and the resulting urea dosing control,
- The particle number filtration efficiency, which is verified at stationary engine operation, is perfectly valid also at the transient operation.

The present results will be confirmed in the further project activities with other systems and with different testing cycles. A special attention will be paid to the operational profiles, which are representative for low emissions zones LEZ.

1. INTRODUCTION

Laboratories for IC-Engines and Exhaust Emission Control of the University of Applied Sciences Biel, Switzerland (AFHB) participate since 1992 at the Swiss activities about nanoparticle analytics and DPF verification.

The upcoming developments of $deNO_x$ (especially SCR) systems and the combinations with DPF's offer a large amount of variants and technical complexity, which represent new challenges not only for the manufacturers, but also for the users and for the responsible authorities.

In the VERTdePN project AFHB collaborates closely with several Swiss specialists of chemistry, catalysis, measuring technics and combisystems (EMPA, PSI, SUVA, ME, UMTEC), as well as European specialist from JRC Ispra, I ; TNO & VROM, NL; AEEDA, B; FAD and TÜV D; AKPF, A.

The application of combined systems (DPF+SCR) as retrofitting raises different technical and commercial problems. In general opinion, this retofitting will be possible mostly through the incentives, or restrictions with respect to low emission zones LEZ, [1] and decisions of several authorities.

The present paper shows the testing procedures of VERTdePN at the current development stage and some examples of results from two very advanced combined retrofitting systems.

2. AVAILABLE TECHNICAL INFORMATION

DPF+SCR

The combination of particle filtration (DPF) and of the most efficient $deNO_x$ technology (SCR) is widely considered as the best solution, up to date, to minimize the emissions of Diesel engines. Intense developments are on the way by the OEM's and a lot of research is performed, [2-16].

The removal of NO_x from lean exhaust gas of Diesel engines (also lean-burn gasoline engines) is a challenge. Selective catalytic reduction (SCR) uses a supplementary substance, a reducing agent, which in presence of catalysts produces useful reactions transforming NO_x in N_2 and H_2O.
The preferred reducing agent for toxicological and safety reasons is a water solution of urea (AdBlue), which due to reaction with water (hydrolysis) and due to thermal decomposition (thermolysis) produces ammonia NH_3, which is the actual reducing substance.

A classical SCR deNO_x system consists of four catalytic parts:

- precatalyst converting NO to NO_2 (with the aim of 50/50 proportion)
- injection of AdBlue (with the intention of best distribution and evaporation in the exhaust gas flow)
- hydrolysis catalyst (production of NH_3)
- selective reduction catalyst (several deNO_x reactions)
- oxidation catalyst (minimizing of NH_3 slip).

The main deNO_x-reactions between NH_3, NO and NO_2 are widely mentioned in the literature. They have different rates depending on the nature of the catalyst, the exhaust temperature, space velocity and stoichiometry of the reducing agent. This offers a complex situation during transient engine operation.
Additionally to that there exists an optimal temperature window for each catalyst and cut off temperature for the AdBlue-injection to prevent the deposits on the catalyst.

Several side reactions can occur forming secondary pollutants. An objective is to minimize the tail pipe emissions of: ammonia NH_3, nitrous oxide N_2O, isocyanic acid HNCO and ammonium nitrate NH_4NO_3 and other secondary nanoparticles, [17-22].

VERT quality testing

VERT was in the 1990's a joint project of occupational insurance agencies from Switzerland (SUVA), from Austria (AUVA) and from Germany (TBG) concerning the reduction of emissions of actual machines in tunnel construction, [23, 24, 25].

It was recognized quickly in the VERT project, that the retrofitting with DPF is the most efficient measure to eliminate radically the particle emissions of Diesel engines in underground. To introduce the DPF-systems for retrofitting it was necessary to establish: the quality criteria and quality test procedure, field control and appropriate support to the users.

One of the most important statements of VERT is, that the validation of filtration efficiency of a DPF by means of particle mass PM (legal parameter up to date) is not sufficient and sometimes misleading. In several cases, particularly with the presence of some catalytic substances in the DPF, sulfates can be produced (only the sulfur from lube oil can be sufficient for that), which pass the DPF as vapor and condensate afterwards on the PM-measuring filter. In an extreme case this can cause, that the DPF, which filters perfectly the solid particles (NP, EC e.g. 98%) seems to double or triple the particle mass (PM).

The filtration efficiency of a DPF can be properly judged only for the solid particles. In this context the nanoparticles are considered in VERT as the most important criterion, [26, 27]. Complementary information is given by a coulometric analysis of elemental carbon (EC) from the collected PM filter residuum.

The nanoparticulates can be measured with different methods and due to the aptitude of penetrating very easily into the living organisms they are regarded as very dangerous for health, [28, 29, 30].

Since 2001 there are discussions in the international legislative gremia about possibilities of introducing the NPs as a legally limited parameter, as recommended by the Particulate Measurement Program (PMP) of the UN Working Party on Pollution and Energy (GRPE), [31, 32, 33].

For some systems, which use catalytic coatings, or fuel additives, or combinations of both of them, a VERT secondary emission test (VSET) has to be performed.

For retrofitting with combined systems (DPF+SCR) quality testing and fulfilment of certain criteria are necessary both: for the user and for the authority.

The Swiss VERT Network started the works to include the deNO$_x$-systems (SCR, EGR, storage catalysts) in the VERT verification procedures (VERT dePN Programm).

3. VERTdePN

Research subjects and objectives

A general objective of VERTdePN is to include the combined DPF+SCR systems in the test procedures, which were previously developed for DPF applications only.

Since the stationary testing of SCR for onroad application will be not sufficient any more, a simplified dynamic test procedure should be found, which nevertheless would be representative for the legal HD transient testing.

Different variants of catalyst and/or their sequences used for different types of SCR systems, different sequences of DPF and SCR, different possibilities of introduction, homogenization and control of urea and finally different applications offer a large multitude of cases, which will be considered during the tests.

For the VERT DPF quality procedure the research objectives were:

- filtration quality
- durability
- control - & auxiliary systems
- secondary emissions.

The new objectives for a SCR system in the VERTdePN tests are:

- NO_x reduction efficiency
- NO_2- and / or NH_3- slip
- Operating temperature window
- dynamic operation
- field application & durability
- auxiliary systems
- further secondary emissions.

The main structure of VERTdePN tests for combined DPF-SCR is similar, as the preceding VERT activities for DPF, **Fig. 1**:

- Quality test and basic investigation on dynamic engine dynamometer on a representative HD-engine,
- Supervised field test 2000h,
- Analyses of toxic and harmful secondary emissions.

When the DPF of the combined system is already approved by VERT, only simplified tests for the SCR-part will be necessary.

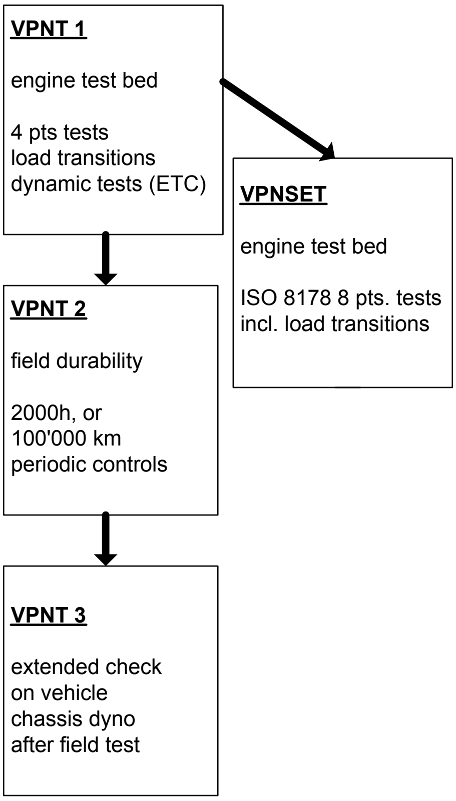


Fig. 1: VERTdePN test procedures for product standards of combined systems (DPF + SCR)

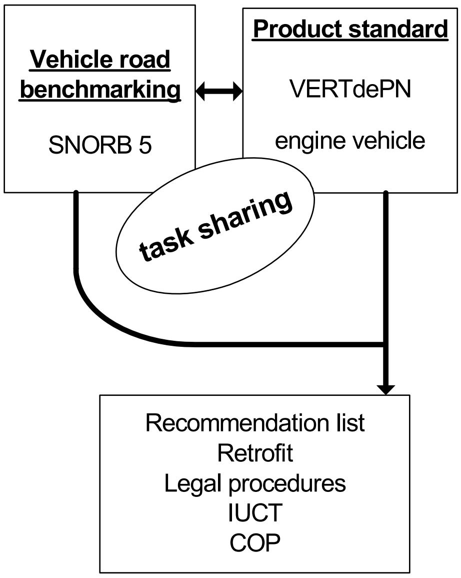


Fig. 2: VERTdePN test procedures for product standards and legal admission of combined systems (DPF + SCR)

Standards for retrofitted vehicles

Important questions about: how to use the product standards from VERTdePN to classify the retrofitted vehicles (e.g. for LEZ's) were raised by the representatives of participating authorities.

The steering committee worked further in several meetings on these problems and elaborated some possible procedures of testing and vehicle admission in Switzerland, see the chart in **Fig. 2**. A complementary on road vehicle testing SNORB (Swiss NO_x Road Benchmarking) was proposed.

It is important to point out, that the expression "vehicle homologation" was replaced by "vehicle benchmarking", since a strict homologation procedure according to the EU-steps would, due to complexity and costs, eliminate the possibility of retrofitting in-use diesel engines with combined deNOx-DPF systems.

In the present state of discussions the following main points can be remarked:

- retrofitting, as a quicker and more efficient measure to reduce consequently the air pollution, makes much sense for the society,
- if any authority wants to support retrofitting it has to do it among others by means of more flexible requirements and procedures; this flexibility can and should be adapted to the different levels of political decisions, **Fig. 3**.
- VERT procedures offered the quality standards, guidelines and choice of systems for DPF-retrofitting,
- VERTdePN proposes the solutions for DPF+SCR retrofitting,
- important elements of the test procedures are the extensive tests of the product on an engine dynamometer connected with different kind of vehicle testing,
- there are three kinds of on road testing proposed:
    - on road real world vehicle benchmarking and comparison with OE vehicles with similar technology (proposed project SNORB to be started during 2008),
    - field test with intermediate and final control on the chassis dynamometer (VPNT2 & VPNT3),
    - simplified acceptance test (vehicle stand still).

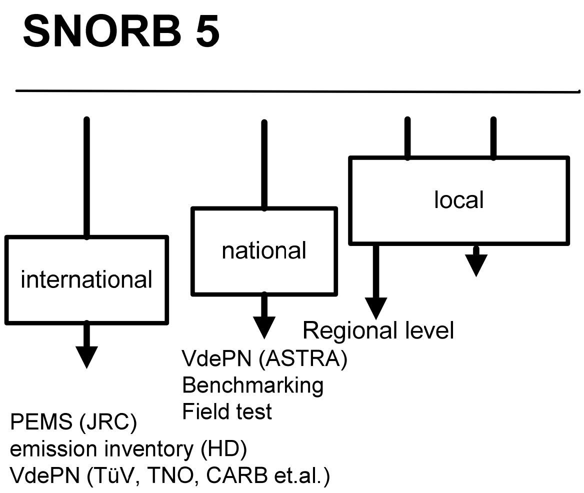


Fig. 3: Swiss NO_x Road Benchmarking EU 5 – validation of retrofitting on different political levels

Further details of these procedures will be elaborated in the coming VERTdePN activities.

4. TEST-ENGINE, FUEL AND LUBRICANT

Test engine

Manufacturer:	*Iveco, Torino Italy*
Type:	*F1C Euro 3*
Displacement:	*7.01 Liters*
Rated RPM:	*max. 4200 rpm*
Rated power:	*100 kW@3500rpm*
Model:	*4 cylinder in-line*

Combustion process:	*direct injection*
Injection system:	*Bosch Common Rail 1600 bar*
Supercharging:	*turbocharger with intercooling*
Emission control:	*none*
Development period:	*until 2000 (Euro 3)*

Fig. 1 shows the engine and the apparatus for nanoparticle analytics SMPS & NanoMet in the laboratory for IC-engines, University of Applied Sciences, Biel-Bienne.

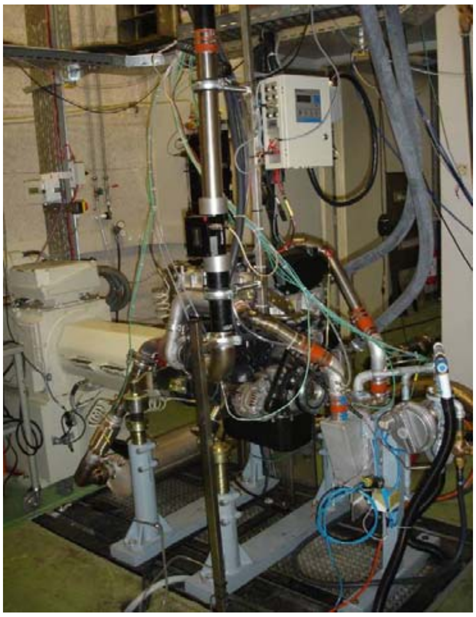


Fig. 4a: IVECO engine F1C with the dynamic dynamometer.

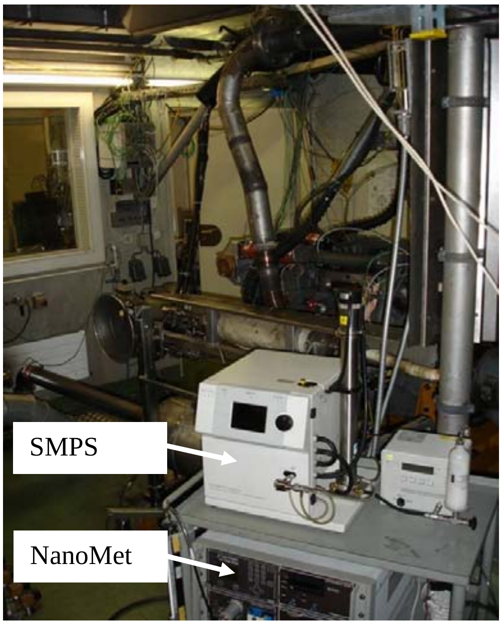


Fig. 4b: Equipment for nanoparticle measurements in the engine room

Fuel

Following fuel was used for the research:

- Shell Formula Diesel fuel Swiss market summer quality (10 ppm S) according to SN EN 590

Lubricant

For all tests a special lubeoil Mobil 1 ESP Formula 5W-30 was used.

Table 2 shows the available data of this oil, ACEA classes: C3, A3, B3/B4, API classes: SL / SM; CF

Table 1 represents the most important data of this fuel according to the standards.

		Diesel
Density at 15°C	g/m	0.842*
Viscosity at 40°C	mm^2/s	2.0 - 4.5
Flash point		above 55°C
Cloud point		max -10°C
Filterability CFPP		max -20°C
Ash	%	max 0.010
Sulfur	ppm	<10
Cetane Number		51
Calorific value	MJ/kg	42.7
C fraction	in %	86.7
H fraction	in %	13.3
O fraction	in %	0
Air / $Fuel_{stoichiom}$	kg/kg	14.52
Boiling range 10-90% °C		180 - 340

Table 1: Fuel properties as per EU-standards (EN)

Property	Mobil	
Viscosity kin 40°C	72.8	mm^2/s
Viscosity kin 100°C	12.1	mm^2/s
Viscosity index	164	(--)
Density 15°C	0.850	kg/m^3
Pourpoint	-45	°C
Flamepoint	254	°C
Total Base Number TBN	14.2	mg KOH/g
Sulfur ashes	6000	mg/kg
Sulfur	7'280	mg/kg
Mg	<10	mg/kg
Zn	1'570	mg/kg
Ca	4'760	mg/kg
P	1'370	mg/kg

Table 2: Data of the applied lubrication oil (EN)

5. MEASURING SET-UP AND INSTRUMENTATION

Engine dynamometer and standard test equipment

Fig. 5 represents the special systems installed on the engine, or in its periphery for analysis of the regulated and unregulated emissions.

Laboratory equipment employed:

- Dynamic test bench Kristl & Seibt with force transducer HBM T10F
- Tornado Software Kristl & Seibt
- Fuel flow measurement AIC 2022
- Air mass meter ABB Sensiflow P
- Pressure transducers Keller KAA-2/8235, PD-4/8236
- Thermo-couples Type K

Test equipment for exhaust gas emissions

Measurement is performed according to the Swiss exhaust gas emissions regulation for heavy duty vehicles (Directive 2005 / 55 / ECE & ISO 8178):

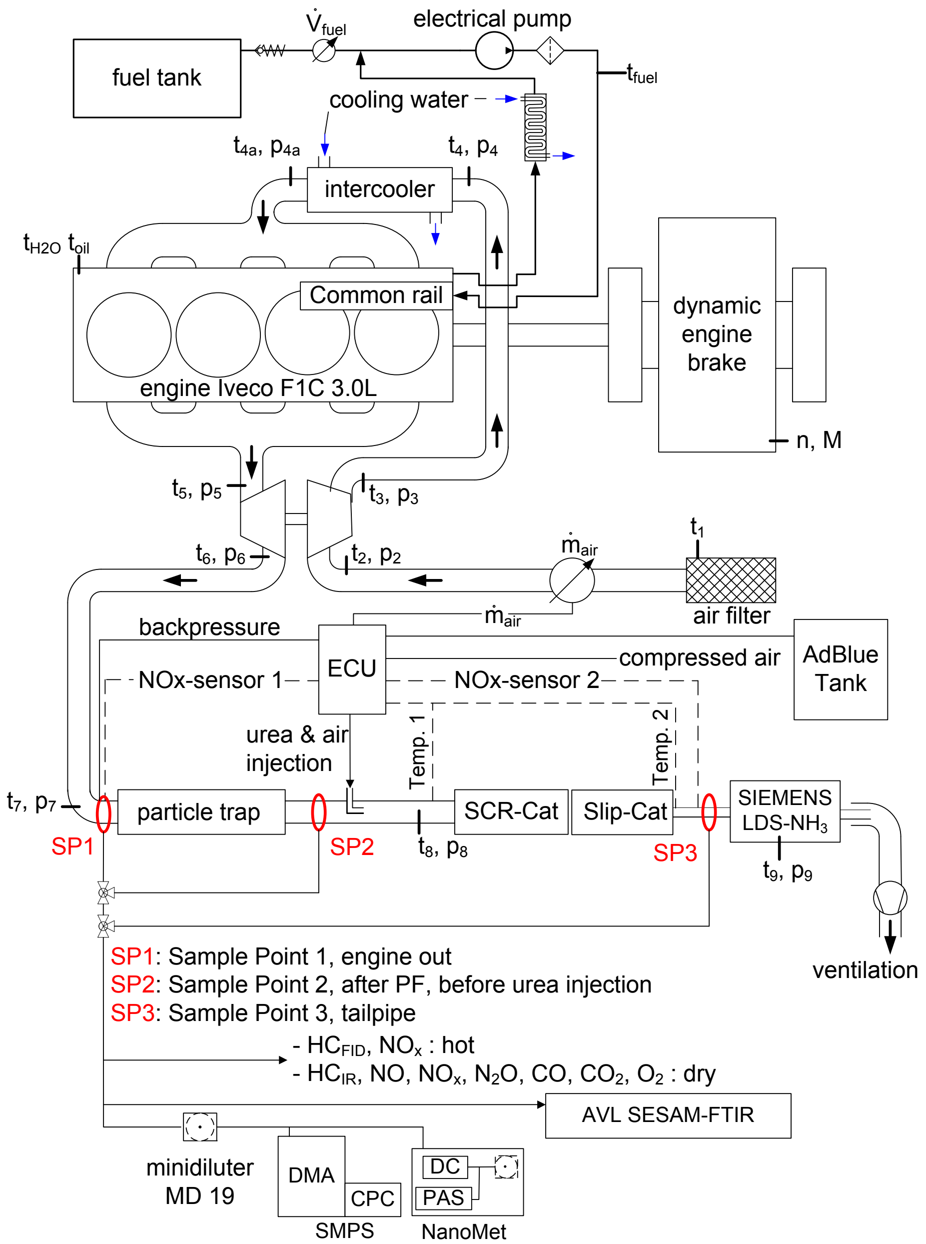


Fig. 5: Engine dynamometer and test equipment

- Volatile components:
  - Horiba exhaust gas measurement devices
    Type: VIA-510 for CO_2, CO, HC_{IR}, O_2,
    Type: CLA-510 for NO, NOx (this standard hot analyser with one reactor is marked in this report as "1 CLD")
  - Amluk exhaust gas measurement device Type: FID 2010 for HC_{FID},

- NH_3 and N_2O:

  With SCR several unregulated and secondary pollutants can be produced. The slip of gaseous components such as ammonia NH_3 and nitrous oxide N_2O was measured by means of:
  - Siemens LDS 6 Laser Analyzer 7MB 6021, NH_3
  - Siemens ULTRAMAT 6E 7MB2121, N_2O
  - Eco physics CLD 822 CM hr with hot line for NO, NO_2, NO_3, NH_3 (this analyzer with two reactors is marked in this report as "2 CLD")

- FTIR (Fourier Transform Infrared) Spectrometer (AVL SESAM) with the possibility of simultaneous, time-resolved measurement of approx. 30 emission components – among those validated are: NO, NO_2, NO_x, NH_3, N_2O.

Particle size analysis

To estimate the filtration efficiency of the DPF, as well as to detect the possible production of secondary nanoparticles, the particle size and number distributions were analysed with following apparatus, **Fig. 4b**:

- SMPS – Scanning Mobility Particle Sizer, TSI (DMA TSI 3071, CPC TSI 3025 A)
- NanoMet – System consisting of:
  - PAS – Photoelectric Aerosol Sensor (Eco Chem PAS 2000)
  - DC – Diffusion Charging Sensor (Matter Eng. LQ1-DC)
  - MD19 tunable minidiluter (Matter Eng. MD19-2E)
  - Thermoconditioner (TC) (i.e. MD19 + postdilution sample heating until 300°C).

The nanoparticle results represented in this paper are obtained with sampling at tail pipe with MD19 and with thermoconditioner (300°C).

The nanoparticulate measurements were performed at constant engine speed (warm) with SMPS and NanoMet. During the dynamic engine operation NanoMet and CPC were used.

6. TEST PROCEDURES

According to the different objectives of the project several test procedures were used.

After analyzing the backpressure of the system in the entire engine operation map it was decided to limit the operation range.

Fig. 6 shows the limited engine map, the 8-point ISO 8178 cycle in this limited map and the 4-point test, used for VPNT1.

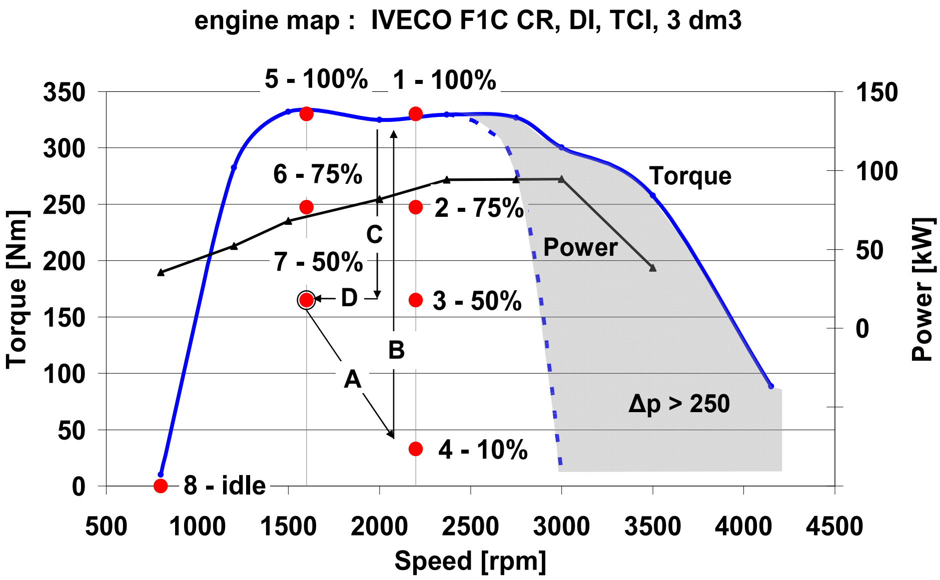


Fig. 6: 8pts. test (ISO 8178) in the limited engine map and setting of the VPNT1 4 pts. test

The 8-points cycle was also used for the secondary emission test VPNSET developed at EMPA. These tests were performed in the present work with three different feed factors α.

For the tests concerning: filtration efficiency, deNO$_x$-rate, unregulated parameters, some basic studies on the investigated systems were performed in the 4-points test according to VPNT1 (AFHB).

These operating points are (in the following sequence):

- operating point 7: 50% load, intermediate speed 1, 1600 rpm / 50%,
- operating point 4: 10% load, intermediate speed 2, 2200 rpm / 10%,
- operating point 1: 100% load, intermediate speed 2, 2200 rpm / 100%,
- operating point 3: 50% load, intermediate speed 2, 2200 rpm / 50%,
- operating point 7: repetition.

The four operating points were chosen in such way, that the switching “off” and “on” of the urea-dosing is included in the tests (pt. 7 → pt. 4 and pt. 4 → pt. 1).

For a more detailed investigation of the tested system different sampling positions (SP) were used (see **Fig. 5**):

SP 0 sampling engine out w/o aftertreatment system
SP 1 sampling engine out with aftertreatment system
SP 2 sampling engine after DPF (before urea dosing) with aftertreatment system
SP 3 sampling engine at tailpipe with aftertreatment system.

This designation of sampling positions is used in the presented figures and in the discussion of results.
The dynamic testing was started with the European Transient Cycle ETC, which was first defined on the basis of the limited engine operation map, **Fig. 7**.

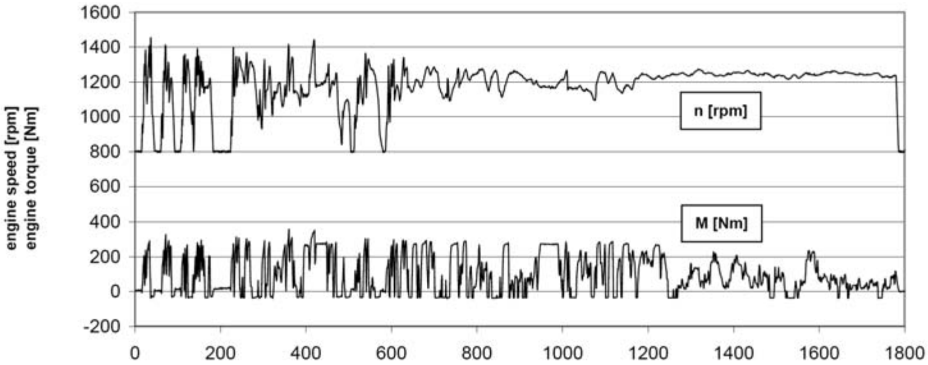


Fig. 7: ETC for the limited version of the engine map, IVECO F1C.

The tests were driven after a warm-up phase, when the engine coolant temperature and lube oil temperature reached their stationary values (stationary points tests).
Before the start of each dynamic cycle the same procedure of conditioning was used to stabilize the thermal conditions of the exhaust gas aftertreatment system. This conditioning was: 5 min at point 1 and 0.5 min of idling.

7. RESULTS

The results were obtained from a combined system consisting of a coated DPF upstream the urea dosing and a SCR catalyst downstream (as in Fig. 5). Sometimes an ammonia slip catalyst was used as a modulus at the end of the system. This (DPF+SCR) system is designed for transient application. It has an electronic control unit, which uses the signals of: air flow, NO_x before/after system and temperatures before/after SCR modulus.

Stationary engine operation

Fig. 8 shows the time-plots of NO_x and NH_3 in the 8-points test with different urea feed factors α. Increasing the feed factor up to α = 1.2 enables a deNO$_x$ efficiency up to 98%, but also increases the ammonia slip up to 125 ppm. **Table 3** illustrates this at one operating point (2200 rpm / 100%). At low load operation (OP 4 & OP 8) there is no urea feeding and consequently no NO_x-reduction.

	w/o DPF + SCR	with DPF + SCR		
2200 rpm / 100%		α = 0.8	α = 1.0	α = 1.2
NO_x 1CLD [ppm]	782.0	159.0	42.0	14.0
RE_{NOX} [%]	-	80.0	95.0	98.0
NH_3 LDS [ppm]	-	6.0	31.0	125.0

Table 3: NO_x reduction efficiency RE & NH_3 depending on feed factor α, (pt. 1 of the 8 pts. test).

Kommentar [nvh1]: One digit less in Table 3

Kommentar [nvh2]:

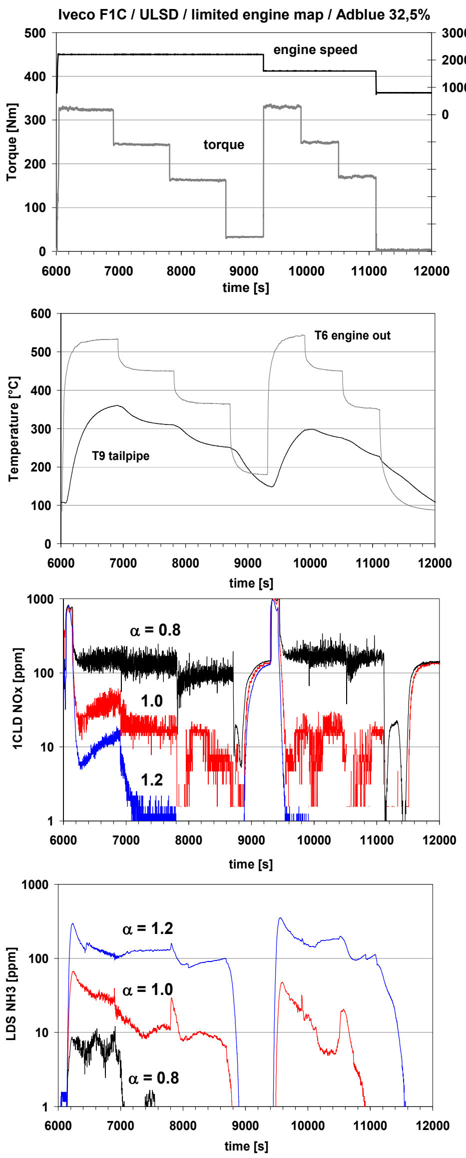


Fig. 8: Comparison of results at 8 points-test with different urea feed factors α

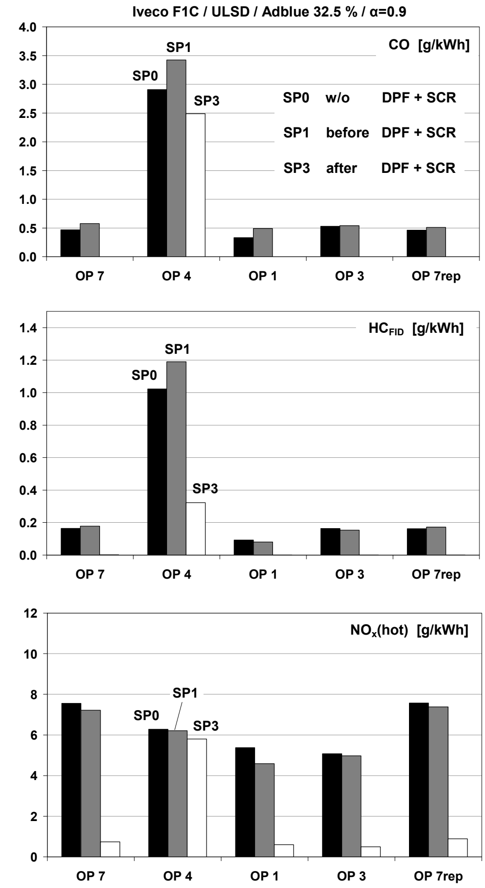


Fig. 9: 4-points test: emissions at different sampling ponts's

Fig. 9 represents emissions at different sampling positions SP in the 4-points test with α = 0.9. There are some differences between CO and HCs at SP0 (without aftertreatment system) and SP1 (before aftertreatment system) caused by a slightly higher backpressure with the installed system.

Due to the use of a catalytic DPF there is an efficient oxidation of CO and HCs between SP1 and SP3, except for the low load operation OP4.

The verification of conversion rates for CO, HC and NO_x as shown in **Fig. 10**, does not show any significant differences, when refering to engine-out emissions with or without aftertreatment system. The maximum stationary ammonia slip at OP1 is 15 ppm.

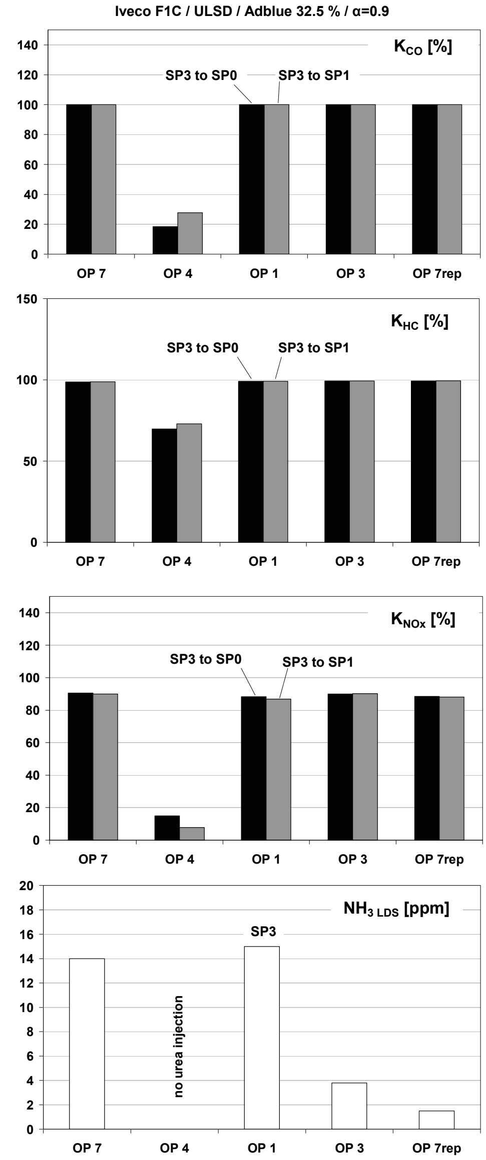


Fig. 10: 4-points test: conversion rates at different SP's and NH_3 tail pipe

Fig. 11 shows the results obtained with FTIR at different sampling positions. Comparing engine-out emissions with those of SP2 (after DPF, before urea dosing) and SP3 (after system).

As expected there is an efficient reduction of nitrogen oxide emissions NO_x, including NO and NO_2 over the SCR catalysts. Exception is at the low load OP4 with no admission of reducing agent.

The production of NO_2 in the catalytic DPF is demonstrated by the emission differences between SP0 and SP2 (**Fig. 11**). At OP4 the exhaust gas temperature is to low and consequently no NO_2 is produced.

N_2O has the tendency to be partialy increased in the DPF and in the SCR, nevertheless, the released quantities are small (<1ppm).

Measurements of nanoparticles NP in the 4-points test at different sampling positions are represented in **Fig.12**. Particularly interesting is the look on the SP2 (after DPF, before urea dosing) and SP3 (after the combined dePN system). There is some production of secondary nanoparticles due to the presence of urea and of other reaction products of deNO_x-chemistry. This is indicated by increased CPC- and DC-values between SP2 and SP3.

The PAS (photoelectric aerosol sensor) is sensitive to the surface of particulates and to the chemical properties of the surface. It indicates solid carbonaceous particles.
The PAS-signals decrease between SP2 and SP3 (Fig. 12), indicating that some of the PAS-active particle surface must be chemicaly changed in the deNOx system.

The DC (diffusion charging sensor) measures the total particle surface independent of the chemical properties of the particles. It indicates both solid particles and condensates.

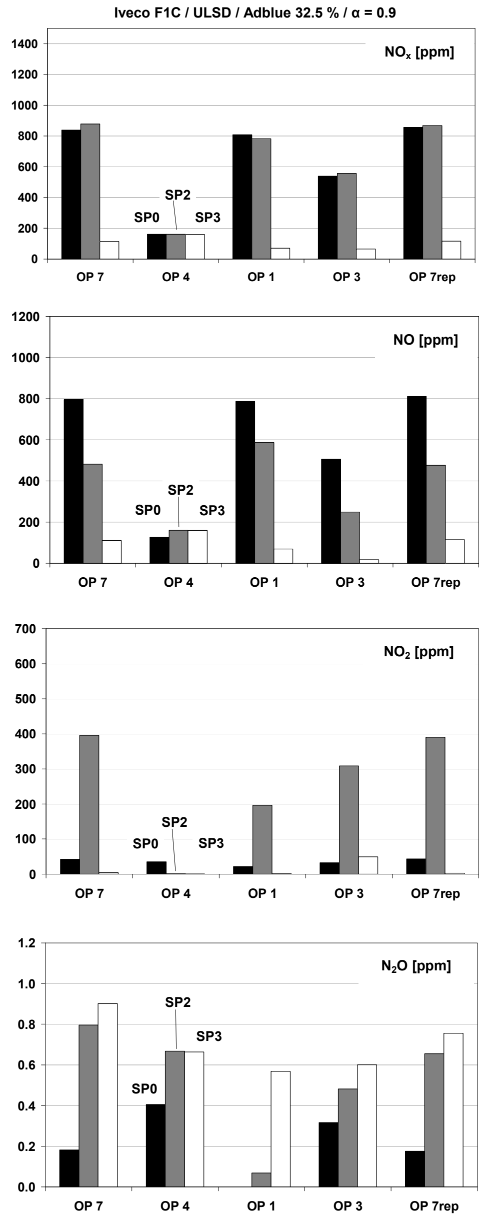


Fig.11: FTIR results in 4-points test at different SP's

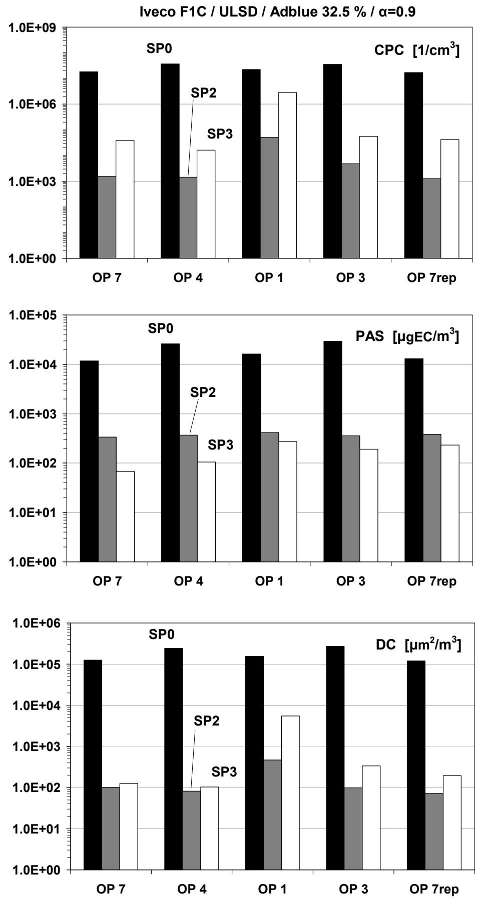


Fig.12: Secondary nanoparticles in the 4-points test (w/o slip cat.)

As known from the literature, secondary pollutants such as cyanuric acid, ammonium nitrate and others can form during the deNOx process. In addition, unreacted urea can also be released. The chemical composition of these secondary aerosols will be studied in further phases of the project.

Kommentar [nvh3]: I do not trust these data and would therefore not publish them at this stage. There is enough data in this article that seems to be very reasonable. Especially the 30'000 ppm of isocyanic acid emissions seem to be very unhealthy.

The increase of NP number concentration (CPC) or of the total surface of the aerosol (DC) over the SCR-system (SP2-SP3) is small compared with the reduction of NP in the DPF (SP0-SP2). Therefore, the secondary formation of nanoparticles does not impact the overall filtration efficiency of the system (notice the logarithmic scale in Fig. 12). Exception is the operating point OP1 with the highest space velocity and an intense secondary formation of nanoparticles.

A summary of reduction efficiencies RE in the 4-points test is represented in **Table 4**. NO_x and NO_2- values of CLD and FTIR, as well as all NP-values (CPC, PAS, DC) are given.
At operating points 7, 1 and 3 the SCR system is working in the optimal temperature window and deNO_x-efficiencies are in the range of 86 - 91%.

	4-Pt. SP3 w/o slip catalyst				4-Pt. SP0 Reference							
$RE_X = \frac{X_{w/o} - X_w}{X_{w/o}} \cdot 100$	with DPF + SCR				without DPF + SCR				RE [%]			
	7	4	1	3	7	4	1	3	7	4	1	3
Temp. T 7 [°C]	339	177	528	367	337	175	490	350				
NO_x 1CLD [ppm]	100	143	85	52	760	140	740	490	87	-2	89	89
NO_x FTIR [ppm]	113.97	160.22	69.86	65.42	838	160	808	539	86	0	91	88
NO_2 1CLD [ppm]	3	1	0	32	40	30	30	30	93	97	100	-7
NO_2 FTIR [ppm]	4	0.56	0.91	49	42	35	21	33	91	98	96	-51
CPC [1/cm^3]	3.9E+04	1.6E+04	2.9E+06	5.6E+04	1.9E+07	3.7E+07	2.2E+07	3.6E+07	100	100	87	100
PAS [µgEC/m^3]	67.6	104.9	274.3	190.2	1.2E+04	2.6E+04	1.6E+04	2.9E+04	99	100	98	99
DC [µm^2/cm^3]	125.4	103.2	5542.1	337.5	1.3E+05	2.4E+05	1.6E+05	2.7E+05	100	100	96	100

Table 4: Integral average values and reduction efficiencies of NO_x, NO_2 and NP in the 4-points test.

Concerning the NO_2 reduction rates there are some open questions: why is the NO_2 efficiency so high at OP4 with no urea dosing and why is it so low at OP3, when the urea injection and temperature range are optimal? These questions can be at least partly explained by the dynamic response of the aftertreatment system in the preceeding, load transitions. There are on the one side the thermal memory effects of the different components (DPF, SCR) in the range of 10 min and on the other side the chemical memory due to store / release effects and secondary reactions.

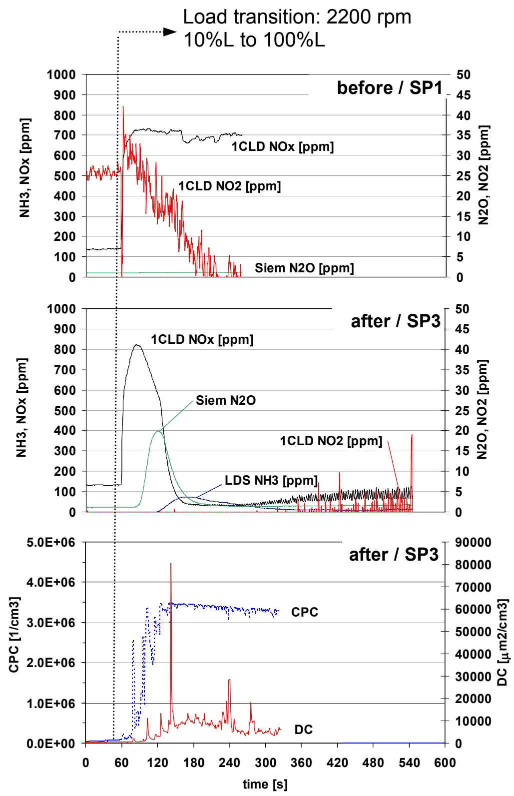
The presented nanoparticle filtration efficiencies (Table 4) are excellent and confirm the required high quality of the DPF part of the system (except of OP1 with highest space velocity and intese secondary NP formation).

Load transitions

The emissions over the time were monitored for transitions A,B,C and D of the 4-points test (Fig. 6). **Fig. 13** shows as an example the transition B with a load increase from 10% (OP4) to 100% (OP1) at 2200 rpm and with urea switching on.

NO_2 levels measured before the combined dePN system (SP1) decline at 100% load, as expected, because of thermal NO_2 decomposition at temperatures up to 490 °C (Table 4).

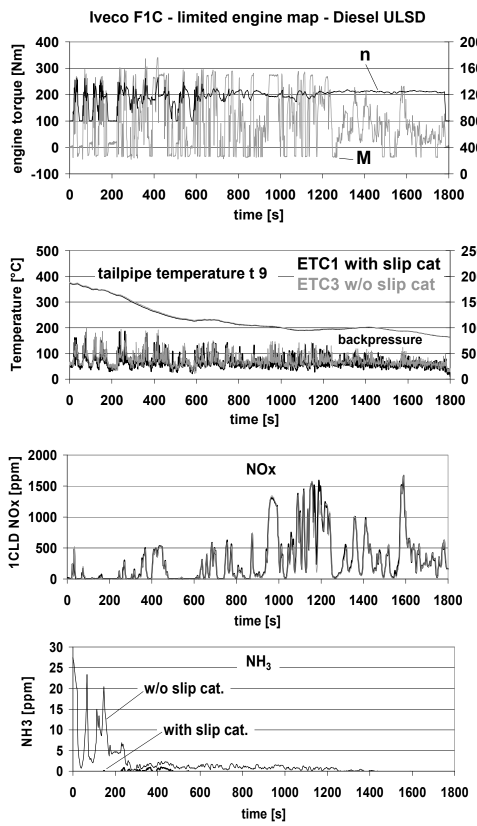
Measured after the system (SP3) quite long response times, in the range of 90 sec, are noticed. In this time, exhaust temperatures increase and the urea dosing starts.



According to the conditions of flow, space velocity, temperature and urea stoichiometry (α) different SCR reactions proceed.
An increase of nanoparticles concentrations is clearly indicated by both, the CPC and the DC.

Load transitions between two stationary engine conditions are very indicative to study in detail the instationary changes in the combined system. Nevertheless for some specific purposes longer operation times at the final stationary state are recommended as well. By extreme load changes (from 0% to 100%) the time necessary for thermal and chemical stabilization of the system can be in the range of up to 20 min.

Fig. 13: Load transition B: from 2200 rpm / 10%L to 2200 rpm / 100%L with measurements before and after DPF + SCR



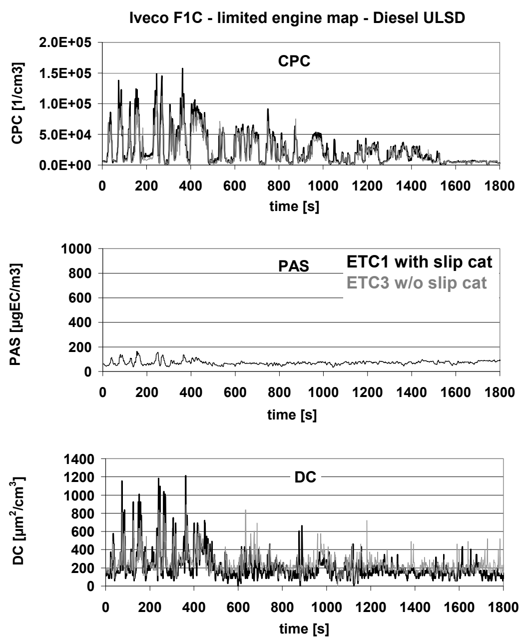


Fig. 14: Comparison of 2 ETCs (ETC1-ETC3), with & w/o slip catalyst at α = 0.9

Dynamic engine operation

These tests were performed in the ETC with limited engine map.
Following results will be shown:
- ETC1 with DPF+SCR+slip cat
- ETC3 with DPF+SCR without slip cat
- ETC4 reference (w/o DPF+SCR).

Before starting each test the thermal condition of the exhaust system was stabilized by repetitive conditioning (see Test Procedures).

Fig. 14 compares emissions during two ETC's with and without slip catalyst. During both tests, exhaust temperatures at the tailpipe decreased below 200 °C and in the second part of the test NO_x emissions increased because of stopped urea dosage..
The ammonia slip catalyst reduced NH_3 emissions, most efficiently in the first phase of the test (until approx. 200 s).

In the first phase of the test (until approx. 500 s) there are also higher emission peaks of NP-emissions CPC & DC, which are an effect of the highly instationary chemistry, production of secondary nanoparticles and store/release phenomena. In the second part of the measuring cycle with less fluctuating engine speed there are also less fluctuations in the CPC- and DC-plots.

Fig. 15 depicts the decreasing NO_x conversion efficiency caused by the cooling of the exhaust system during the test and the respective shut-off of urea dosage. It can be concluded, that with better insulation of the exhaust system, or placing the dePN system closer to the engine, or extending the engine operation range, the deNO_x reduction rates can be influenced. Some of these measures will be tested in further works.

The results of target emission were integrated for different test periods:
- initial period 0-400 s
- final period 1400-1800 s
- overall test 0-1800 s.

The obtained average emission concentration and the reduction efficiencies are summarized in **Table 5**.

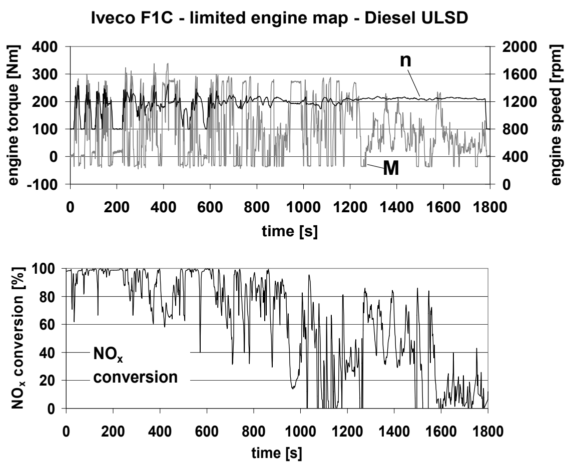


Fig. 15: Comparison of 2 ETC's (ETC3-ETC4), reference & w/o slip catalyst at α = 0.9

Formatiert: Englisch (Großbritannien)

The NO_x- and NO_2-conversion rates decrease during the test, as previously discussed. The NO_x concentrations obtained from CLD and FTIR correspond very well. NO_2 levels are rather low, therefore discrepancies are larger
Again, very high filtration efficiencies of 99 - 100% were noticed despite of some secondary NP-formation in all periods of the ETC.

Kommentar [nvh4]: To many digits

$RE_X = \frac{X_{w/o} - X_w}{X_{w/o}} \cdot 100$	ETC 3 w/o slip catalyst with DPF + SCR			ETC 4 Reference without DPF + SCR			RE [%]		
	0-400 s	1400-1800 s	0-1800 s	0-400 s	1400- 1800 s	0-1800 s	0-400s	1400-1800 s	0-1800 s
Temp. T 7 [°C]	278	197	259	271	195	255			
NO_x 1CLD [ppm]	47	390.9	290.1	605.9	504.7	699.8	92	23	59
NO_x FTIR [ppm]	53	426.2	317	660.8	550.5	759.7	92	23	58
NO_2 1CLD [ppm]	3	42.6	35.9	39	49.2	48.6	92	13	26
NO_2 FTIR [ppm]	2.4	48.5	38	29.2	53.21	40.4	92	9	6
CPC [$1/cm^3$]	3.3E+04	7.6E+03	2.1E+04	4.5E+06	5.9E+06	6.2E+06	99	100	100
PAS [$\mu gEC/m^3$]	65.9	65.4	60.9	6734.8	8638.5	8635.6	99	99	99
DC [$\mu m^2/cm^3$]	293.8	200.1	243.3	37460.6	41075.4	45724.5	99	100	99

Table 5: Average concentrations and reduction efficiencies of NO_x, NO_2 and nanoparticle emissions in different parts of the ETC.

Fig. 16 compares filtration efficiencies of the combined dePN system (DPF+SCR) in stationary and in dynamic engine operation.

In the operating point OP1 of the stationary 4-points test, the influen-ce of the secondary formation of nanoparticles is visible. In the dynamic test, such effects are hardly detectable, due to over-lapping and blurring of all transient effects.

In the dynamic ETC test, the DPF which fullfills VERT quality standards, is as efficient as in stationary tests. Moreover, in stationary testing it is possible to observe phenomena, which are not visible in the transient tests. Such effects can be: storage/ release of sulfates in the exhaust system, influences of additive-particles, or secondary SCR nanoparticles. The stationary testing of DPFs according to the VERT procedures can be confirmed as the best solution.

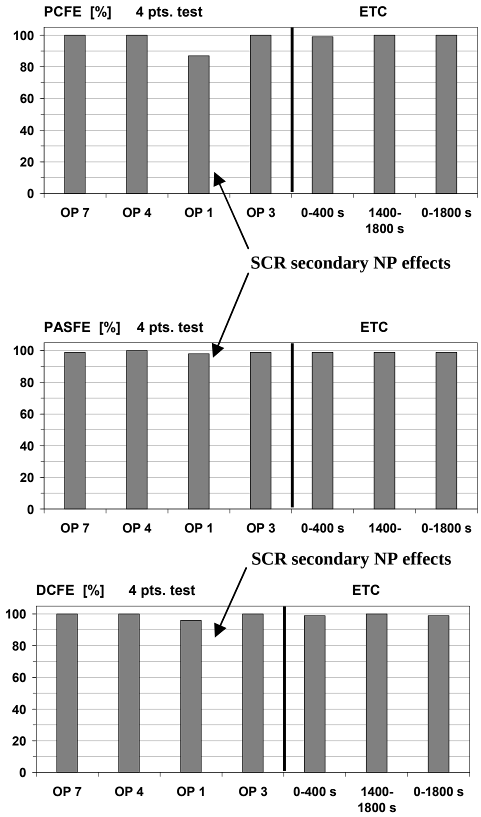


Fig. 16: Filtration efficiencies of the combisystem after SCR – catalyst in stationary and dynamic engine operation

8. CONCLUSIONS

The most important results from the investigated combined DPF+SCR system for transient applications can be summarized as follows:

- the combined dePN systems (DPF+SCR) at transient engine operation efficiently reduce the target emissions with $deNO_x$-efficiencies up to 92% (if operated in the right temperature window) and particle number filtration efficiencies up to 100%,
- with increasing feed factor (up to overstoichiometric urea dosing) NO_x conversion efficiencies increase (up to 98%), but also the ammonia slip rises up to 125 ppm,
- with the recommended feed factor α = 0.9, without slip catalyst, and there is only a moderate average slip of ammonia up to 7 ppm in the ETC and there is a release of small amounts of nitrous oxide of up to 3 ppm,
- the ammonia slip can be efficiently eliminated by a slip-cat,
- during transients there are temporarily increased emissions of nitrogen-containing components, due to momentary imbalanced deNOx reactions,
- in the investigated configuration with urea dosing after the DPF, a secondary formation of nanoparticles is detectable, however with little impact on total number concentrations and overall filtration efficiency of the system,
- the average NO_x conversion efficiency at transient operation (ETC) strongly depends on the exhaust temperatures which are correlated with the urea-dosing strategy,
- the nanoparticle filtration efficiency, which is verified at stationary engine operation, is perfectly valid also at transient engine operation.

The present results will be confirmed in the further project activities with other systems and with different testing cycles. A special attention will be paid to the operational profiles, which are representative for low emissions zones LEZ.

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11. ABBREVIATIONS

AEEDA	Association Europeenne d’Experts en Dépollution des Automobiles
AFHB	Abgasprüfstelle FH Biel, CH
AKPF	Arbeitskreis der Partikelfilterhersteller
	Air min stoichiometric air requirement
ASTM	American Society for Testing Materials
ASTRA	Amt für Strassen, CH, Swiss Road Authority
AUVA	Austria Unfall Versicherung-Anstalt
BAFU	Bundesamt für Umwelt, CH (Swiss EPA)
CARB	Californian Air Resources Board
CDI	Common Rail Diesel Injection
CFPP	cold filter plugging point
CLD	chemoluminescence detector
CNC	condensation nuclei counter
COP	conformity of production
CPC	condensation particle counter
DC	Diffusion Charging Sensor
dePN	de Particles + deNO$_x$
DI	Direct Injection
DMA	differential mobility analyzer
DPF	Diesel Particle Filter
ECU	electronic control unit

ELPI	electric low pressure impactor
EMPA	Eidgenössische Material Prüf- und Forschungsanstalt
EPA	Environmental Protection Agency
ETC	European Transient Cycle
FAD	Förderkreis Abgasnachbehandlungs-technologien für Dieselmotoren, Dresden
FBC	fuel borne catalyst (regeneration additive)
FE	filtration efficiency
FID	flame ionization detector
FTIR	Fourrier Transform Infrared Spectrometer
GRPE	UN Groupe of Rapporteurs Pollution & Energie
HD	heavy duty
ICE	internal combustion engines
IUCT	in use compliance test
JRC	EU Joint Research Center
LDS	Laser Diode Spectrometer (for NH_3)
LEZ	low emission zones
LRV	Luftreinhalteverordnung
ME	Matter Engineering
MD19	heated minidiluter
NanoMet	NanoMetnanoparticle summary surface analyser (PAS + DC + MD19) PAS + DC + sampling & dilution unit
NP	nanoparticles < 999 nm (SMPS range)
OEM	original equipment manufacturer
OP	operating point
PAS	Photoelectric Aerosol Sensor
PC	particle counts
PCFE	particle counts filtration efficiency
PM	particulate matter, particle mass
PMFE	particle mass filtration efficiency
PMP	Particulate Measurement Program of GRPE
PSD	particle size distribution
PSI	Paul Scherrer Institute
RD	relative difference
RE	reduction efficiency
SCR	selective catalytic reduction
SMPS	Scanning Mobility Particle Sizer
SNORB	Swiss NO Retrofit Benchmark
SP	sampling position
SUVA	Schweiz. Unfallversicherungs-Anstalt
TBG	Tiefbaugenossenschaft
TC	thermoconditioner. Total Carbon
TNO	Netherland National, Laboratories
TÜV	Technischer Überwachungsverein, D
ULSD	ultra low sulfur Diesel
UMTEC	Umwelttechnik Institut FH Rapperswil, CH
US-EPA	US – Environmental Protection Agency
VERT	Verminderung der Emissionen von Realmaschinen in Tunelbau
VERTdePN	VERT DPF + VERT deNO_x
VPNT1	VERTdePN Test 1 - engine dyno
VPNT2	VERTdePN Test 2 - field durability 2000h
VPNT3	VERTdePN Test 3 - check after field test chassis dyno
VPNTSET	VERTdePN secondary emissions test - engine dyno
VROM	Netherlands EPA
VSET	VERT Secondary Emissions Test

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IC-Engines and Exhaust Gas Control

VERTdePN – quality verification of combined DPF+SCR systems for retrofitting.

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Dr. Norbert Heeb, EMPA
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Dipl. Ing. Giovanni D'Urbano, FOEN
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NPC ETH Zürich, 23-25. June 2008

Network-Project „VERT dePN-Verification for HD-Retrofitting with combined systems DPF+SCR“

Working laboratories :

AFHB, ME, EMPA, SUVA, UMTEC

Leading:: TTM, AFHB, BAFU

Financial support

BAFU
SUVA
ASTRA

Working packages

Engine	Field
Partner 1	Partner 1
Partner 2	Partner 2
Partner 3	Partner 3

Industrial partners

HUG,
UMTEC Technologie
DINEX,
EMINOX,
HJS,
Johnson Matthey

Open for further partners

Collaborations :

FAD, AKPF, AECC
CARB
SWRI
TNO / VROM

Consulting :

PSI (CCEM, NEADS)
EMPA
LAV-ETHZ
VITO, Belgium
AVL MTC, Sweden

VERTdePN test procedures for product standards of combined systems (DPF + SCR)

VPNT 1

engine test bed

4 pts tests
load transitions
dynamic tests (ETC)

VPNT 2

field durability

2000h, or
100'000 km
periodic controls

VPNT 3

extended check
on vehicle
chassis dyno
after field test

VPNSET

engine test bed

ISO 8178 8 pts. tests
incl. load transitions

VERTdePN test procedures for product standards and legal admission of combined systems (DPF + SCR)

Vehicle road benchmarking

SNORB 5

Product standard

VERTdePN

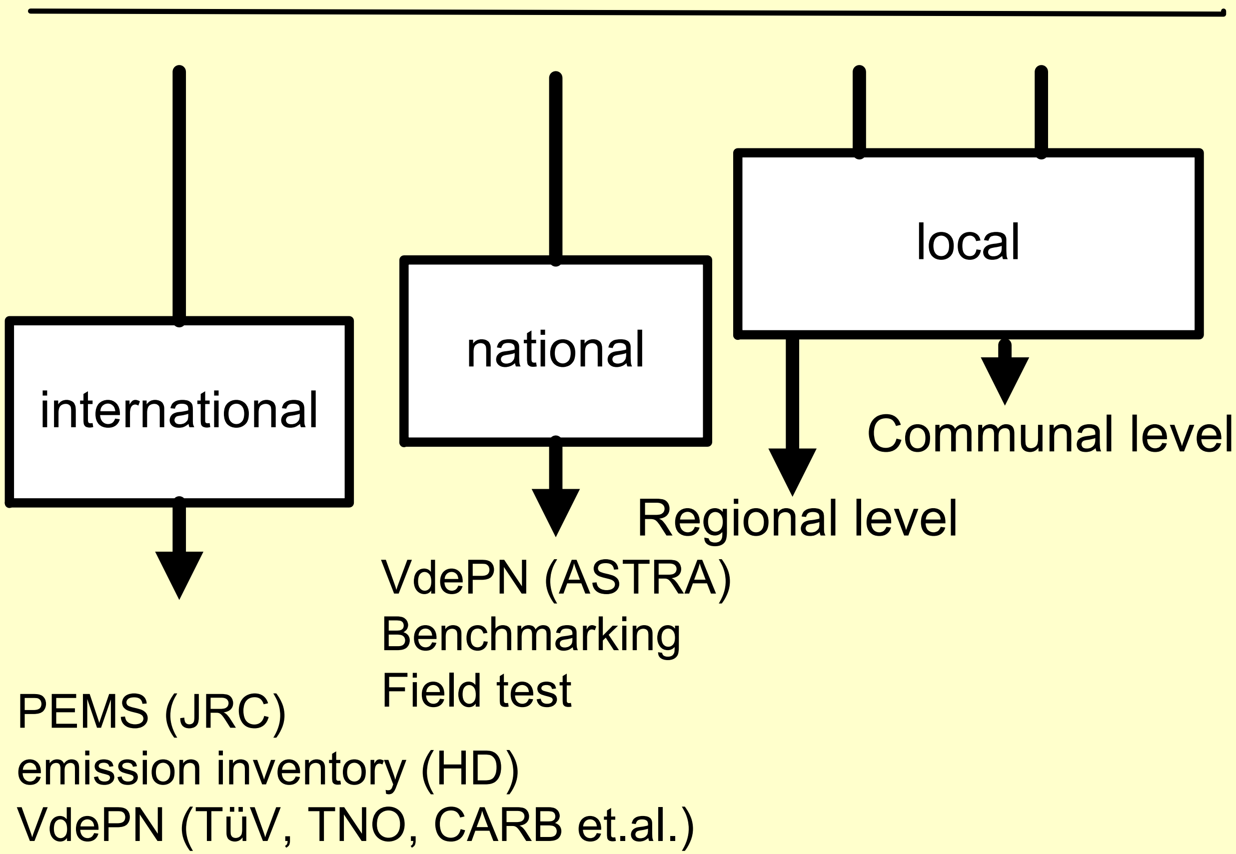
engine vehicle

task sharing

Recommendation list
Retrofit
Legal procedures
IUCT
COP

Swiss NOx Road Benchmarking EU 5 – validation of retrofitting on different political levels

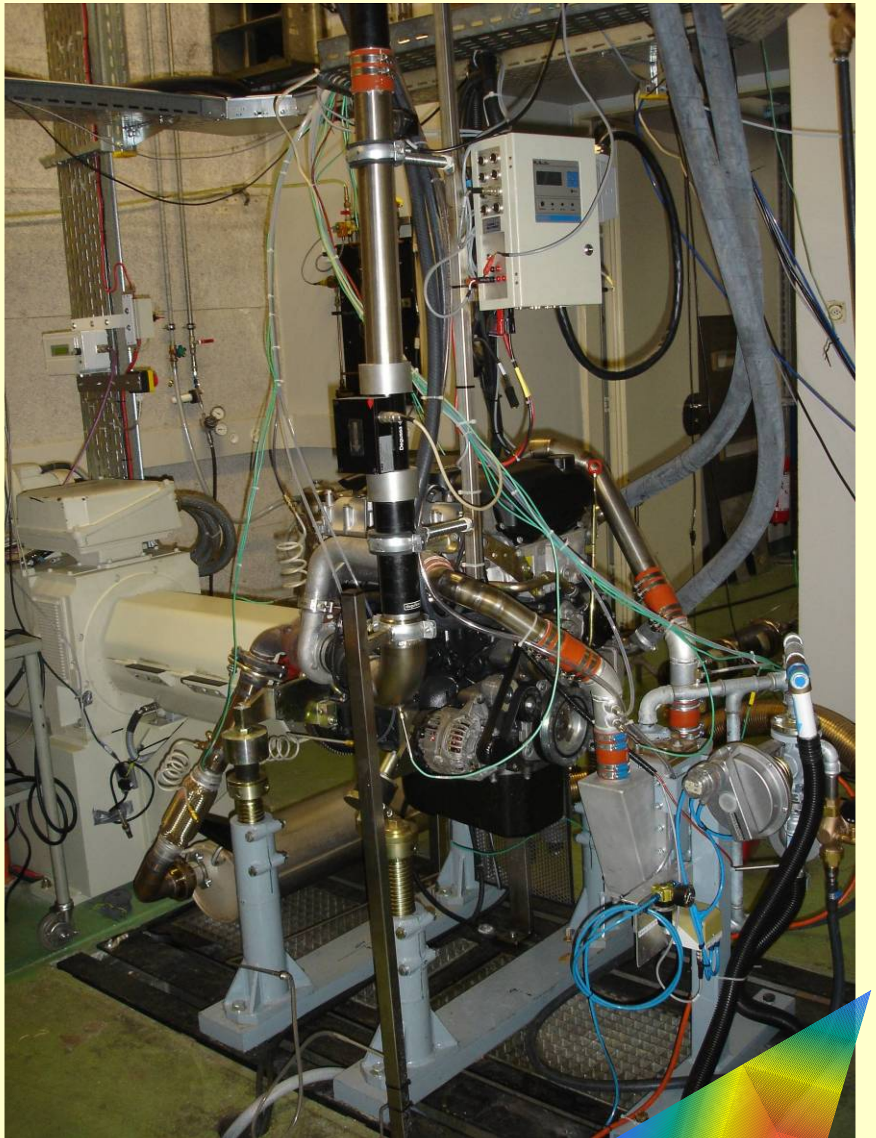
SNORB 5



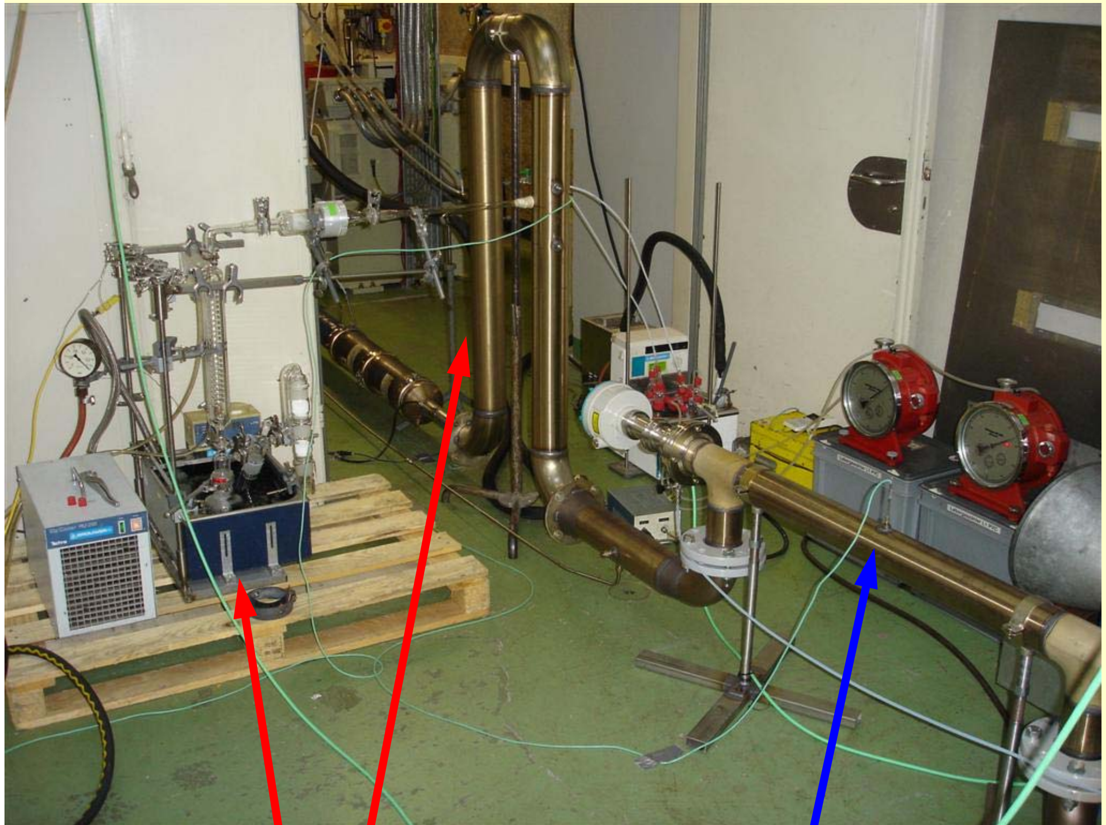
Measuring Set-up

TEST ENGINE

Manufacturer:	**Iveco, Torino Italy**
Type:	**F1C Euro3**
Cylinder volume:	**3.00 Liters**
Rated RPM:	**3500 min^{-1}**
Rated power:	**100 kW**
Model:	**4 cylinder in-line**
Combustion process:	**direct injection**
Injection system:	**Bosch Common Rail**
Supercharging:	**Turbocharger with intercooling**

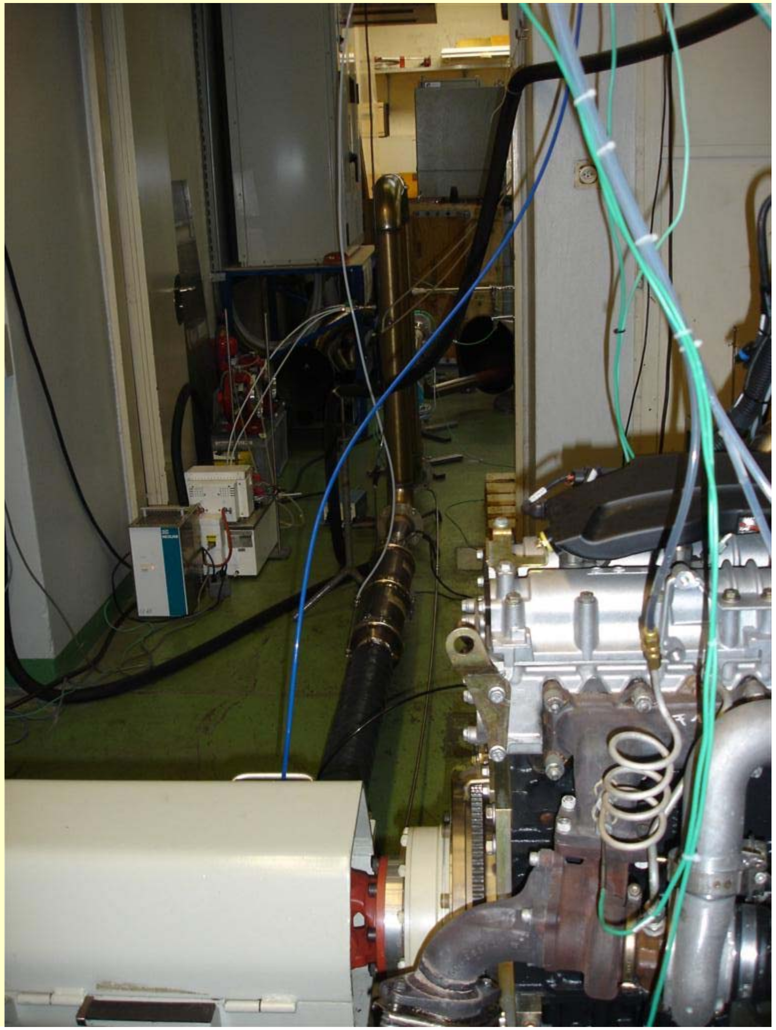


Exhaust line prepared for adaptation of dePN-systems and special exhaust gas analysis

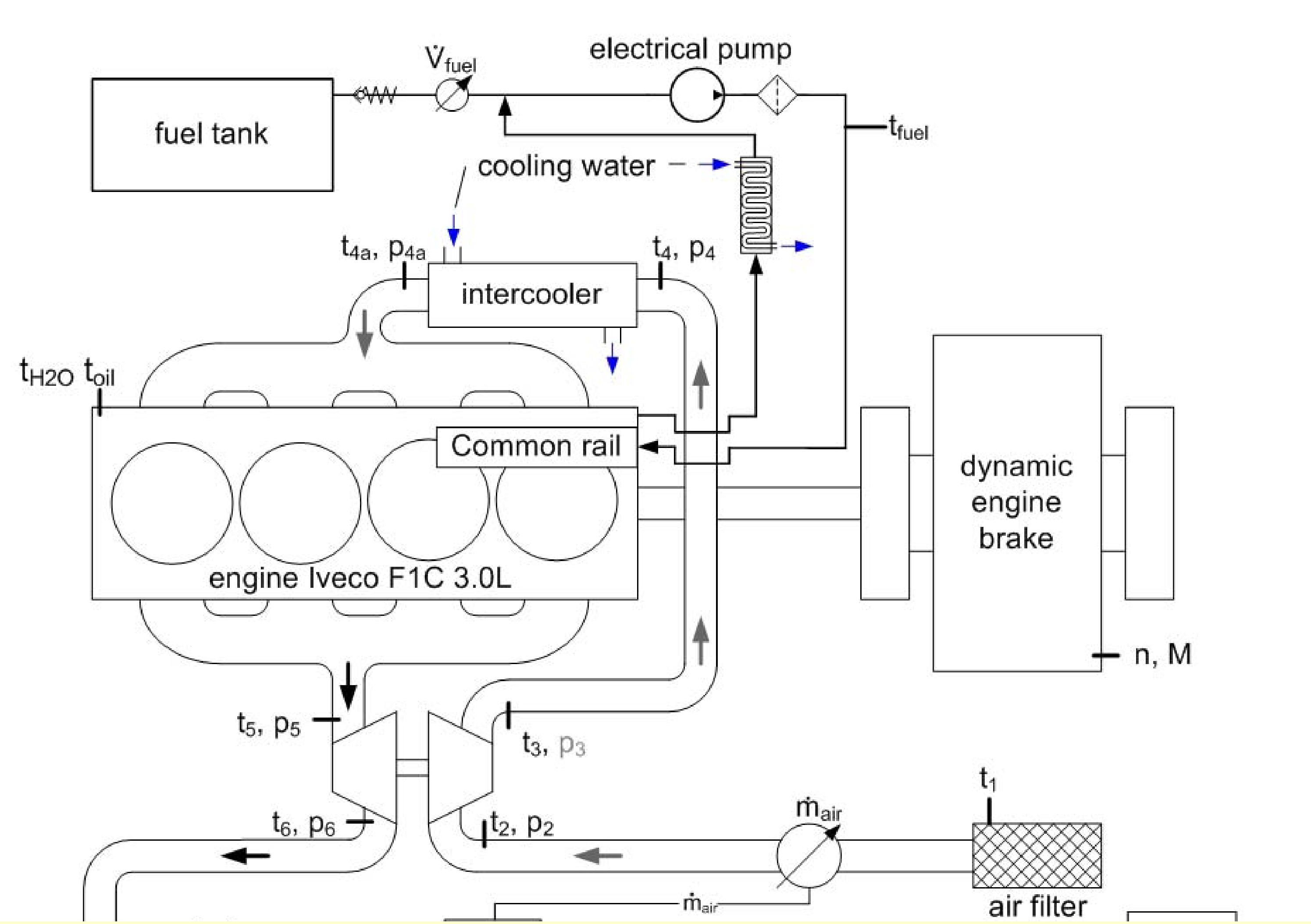


DPNSET-Sampling

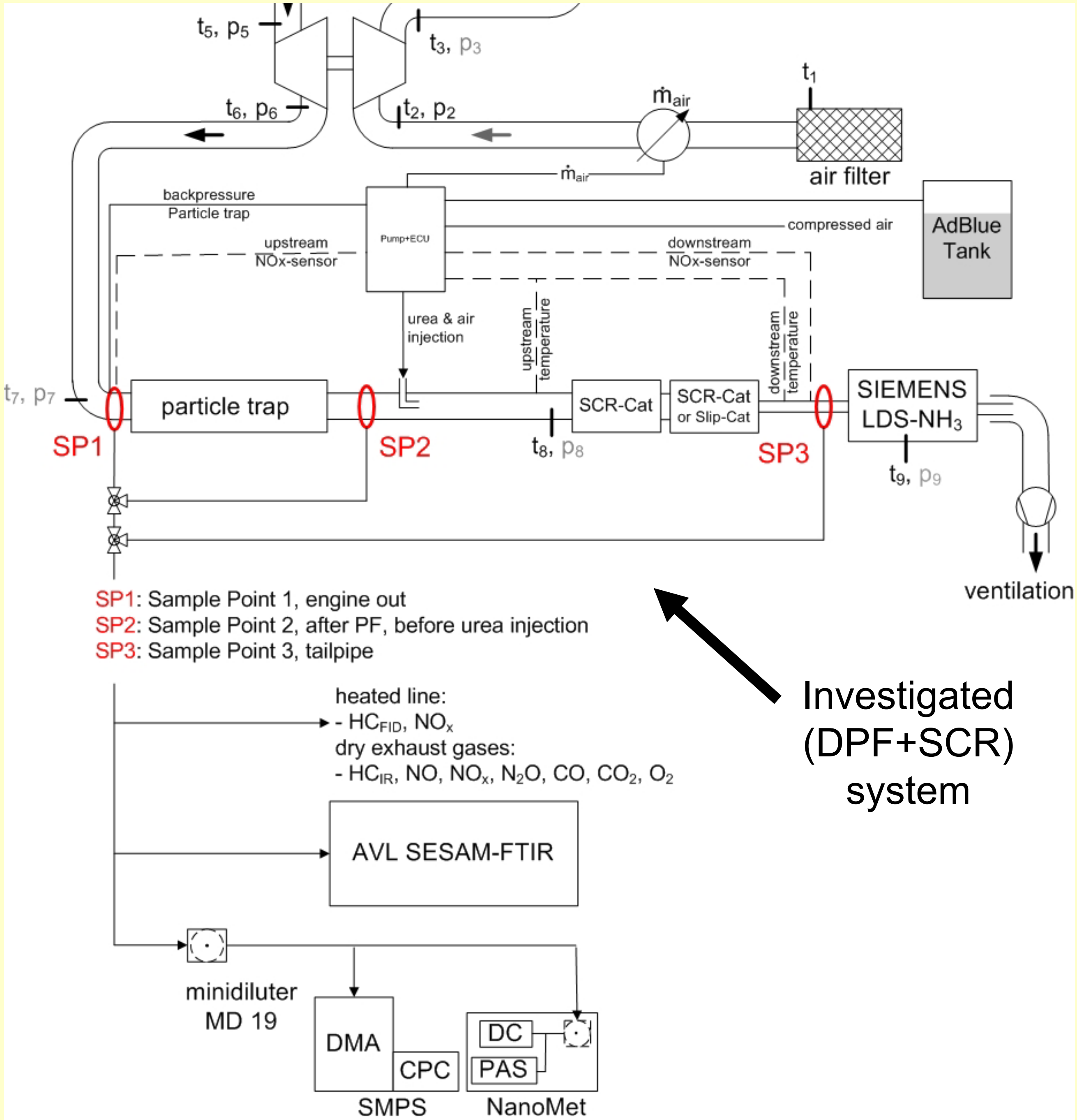
LDS



Measuring set-up (1)



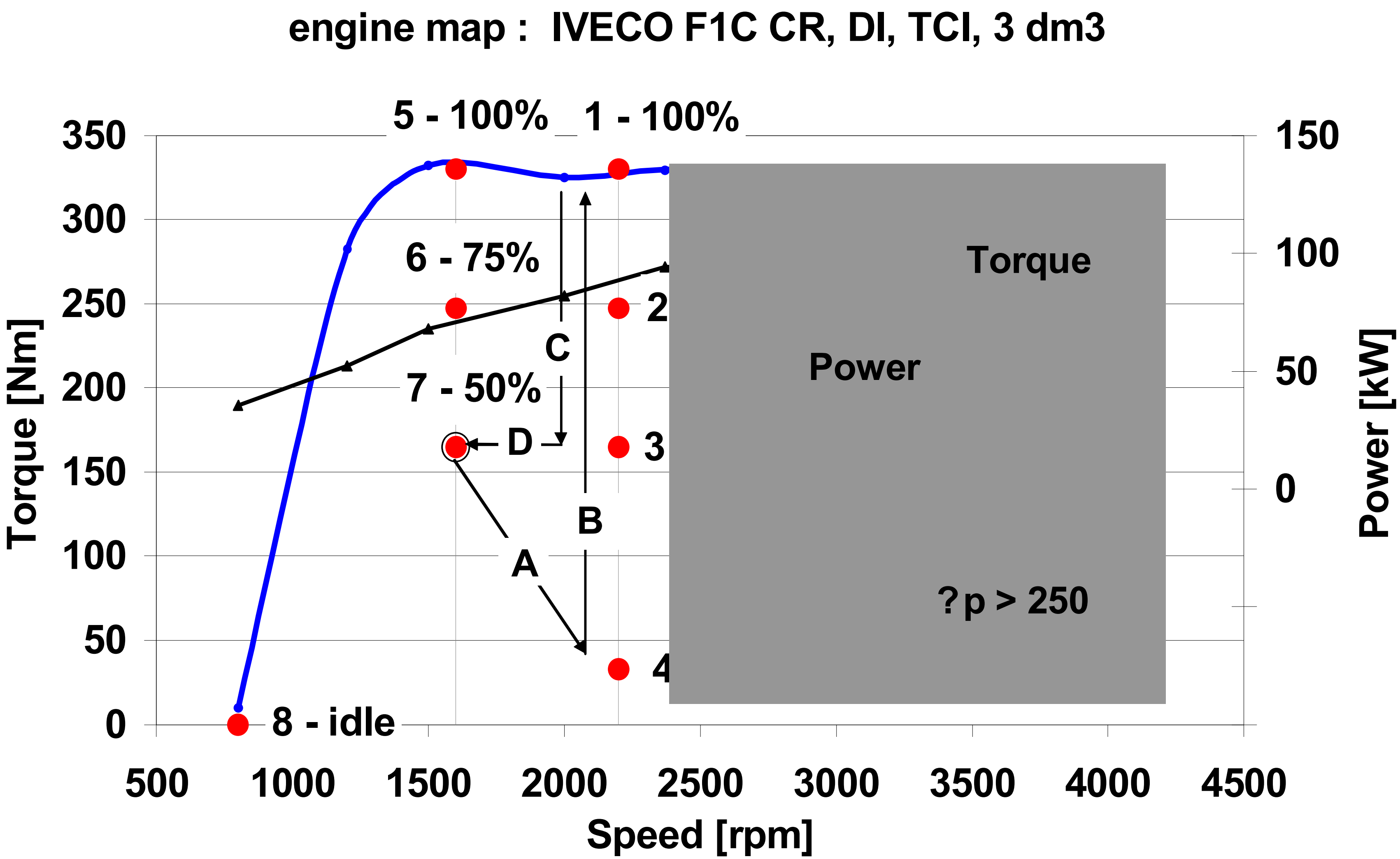
Measuring set-up (2)



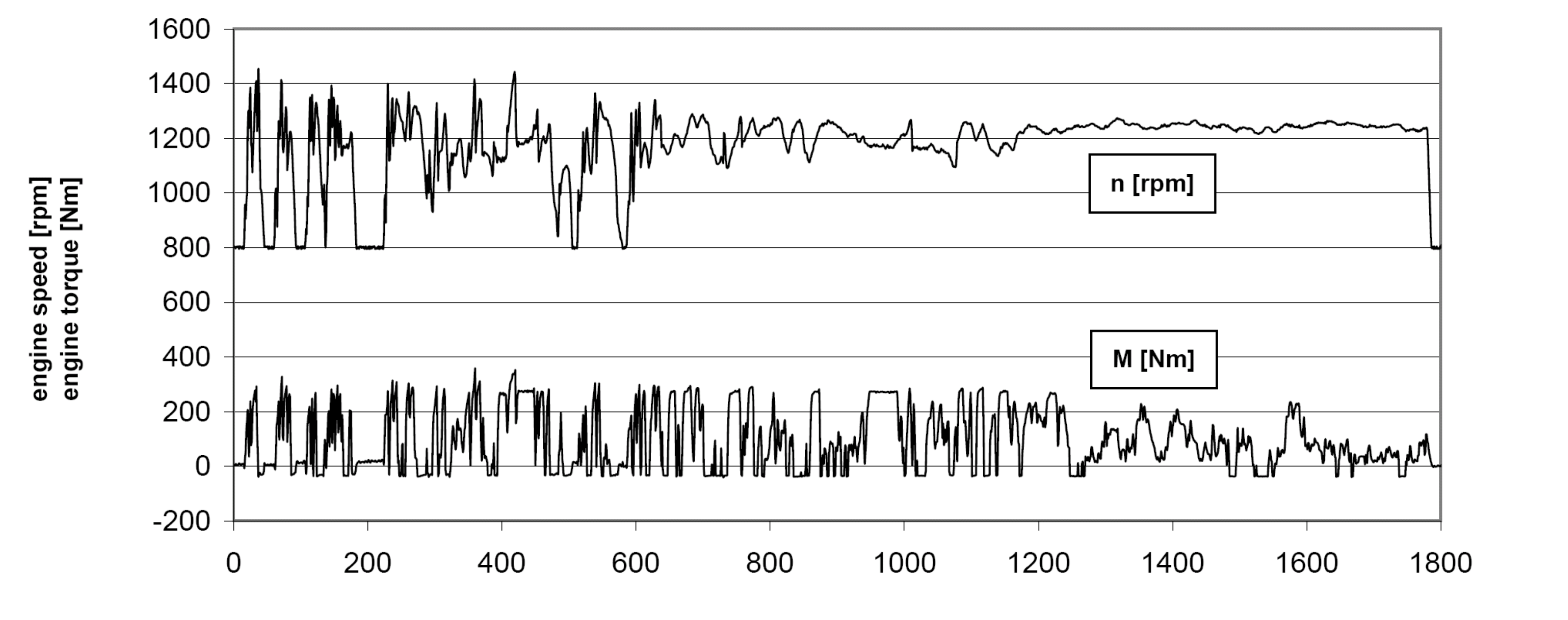
Investigated (DPF+SCR) system

Test Procedures

8pts. test (ISO 8178) and the VPNT1 4pts. test



ETC for the limited version of engine map, IVECO F1C

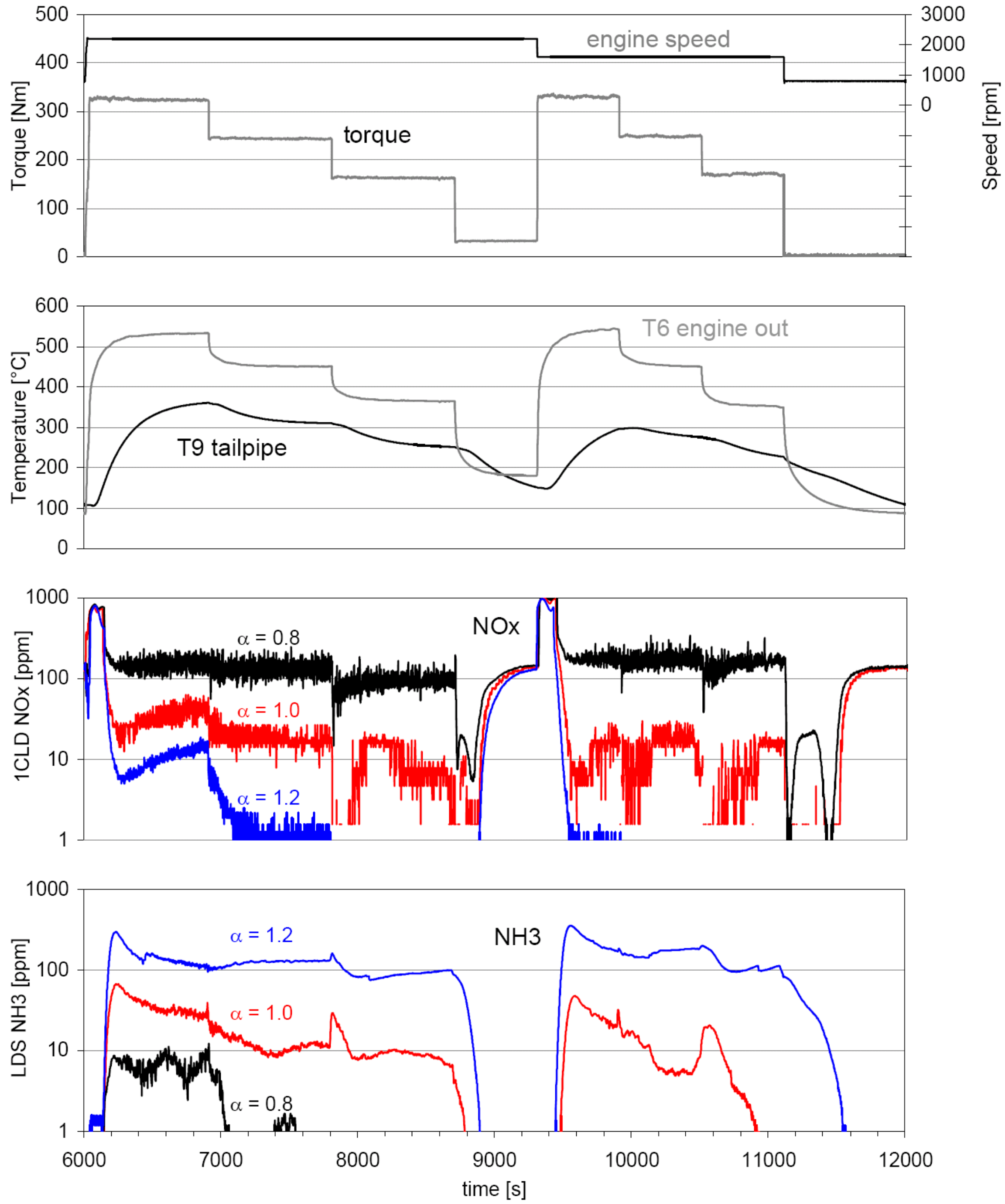


Stationary

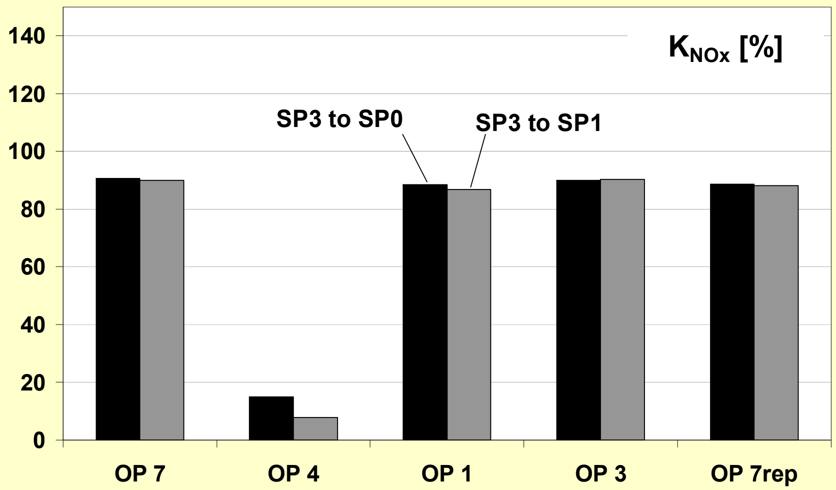
engine operation

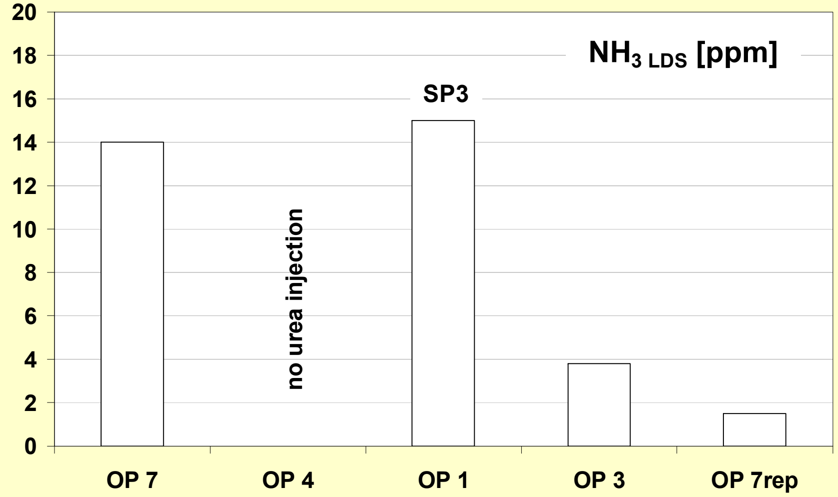
8-points test

Comparison of results with different α

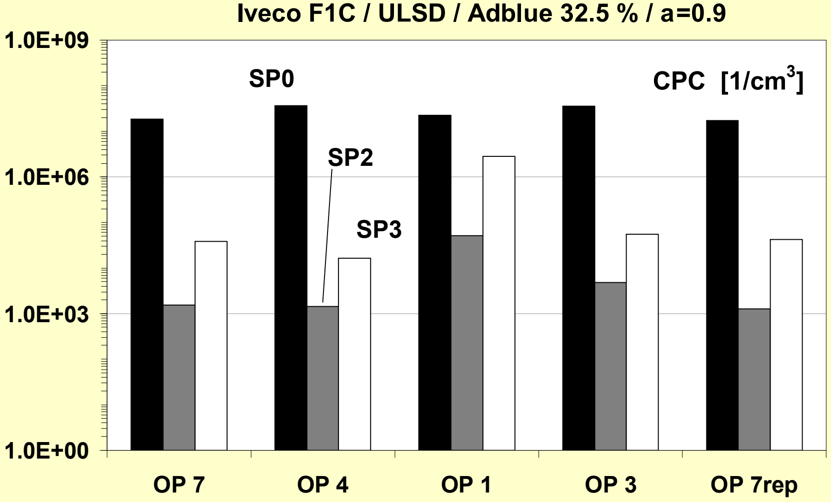


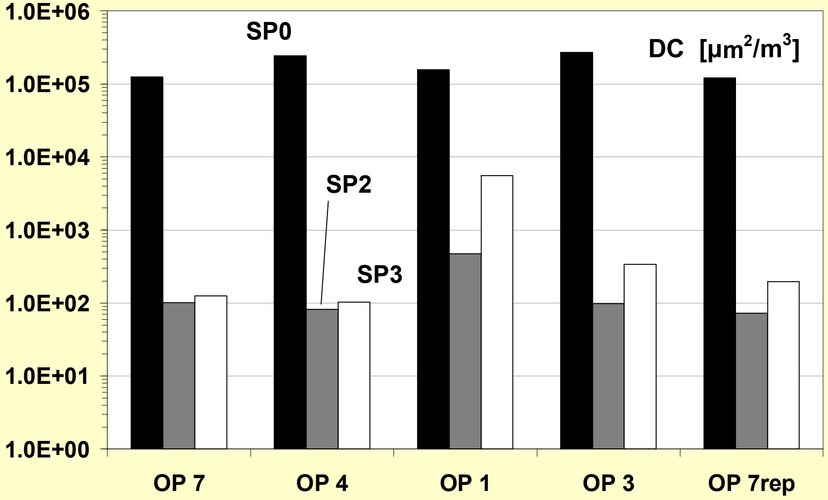
NO_x conversion at different SP's & NH3 tail pipe

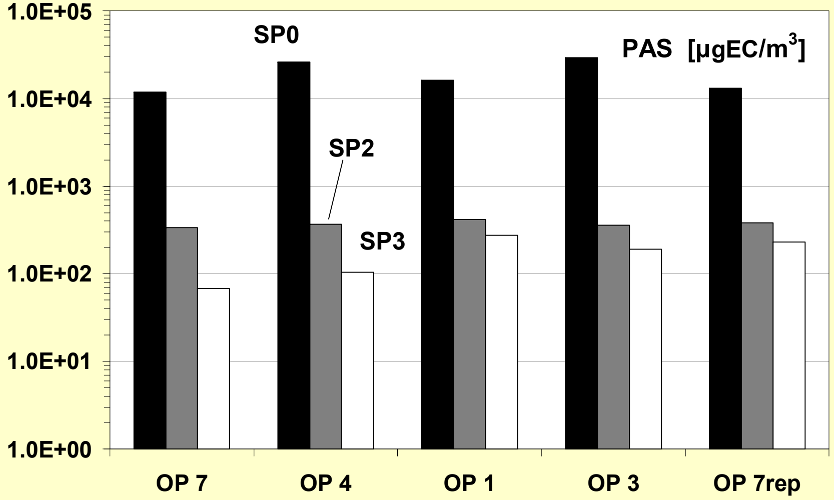




Secondary nanoparticles at 4pts. test (w/o slip cat.)



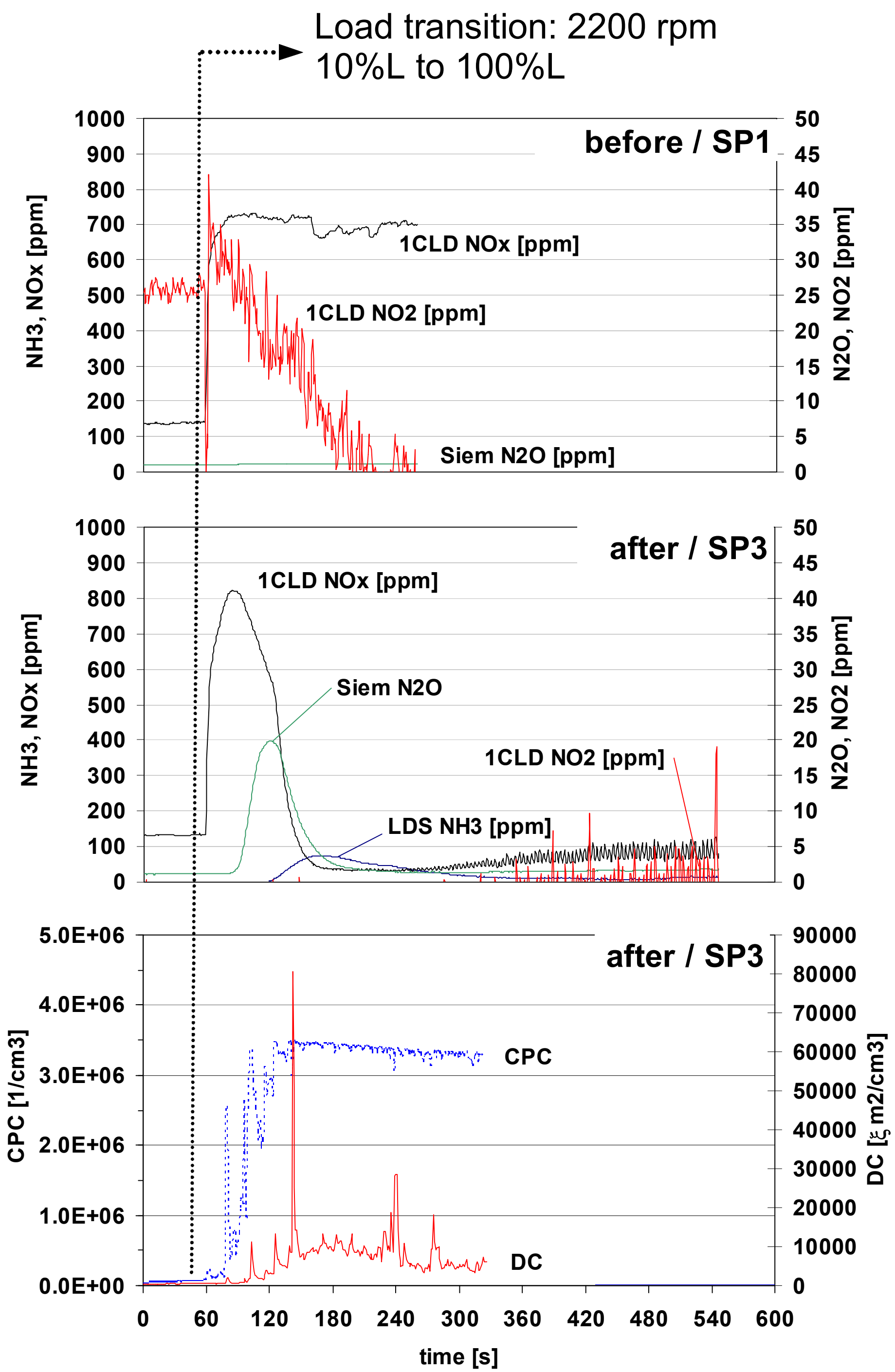




Quasi dynamic

engine operation

→ Laod transitions



Adblue injection switch on

**Load transition B:
from 2200 rpm / 10%L
to 2200 rpm / 100%L
with measurements
before and after
DPF + SCR**

Dynamic

engine operation

→ ETC

ETC's with limited engine map

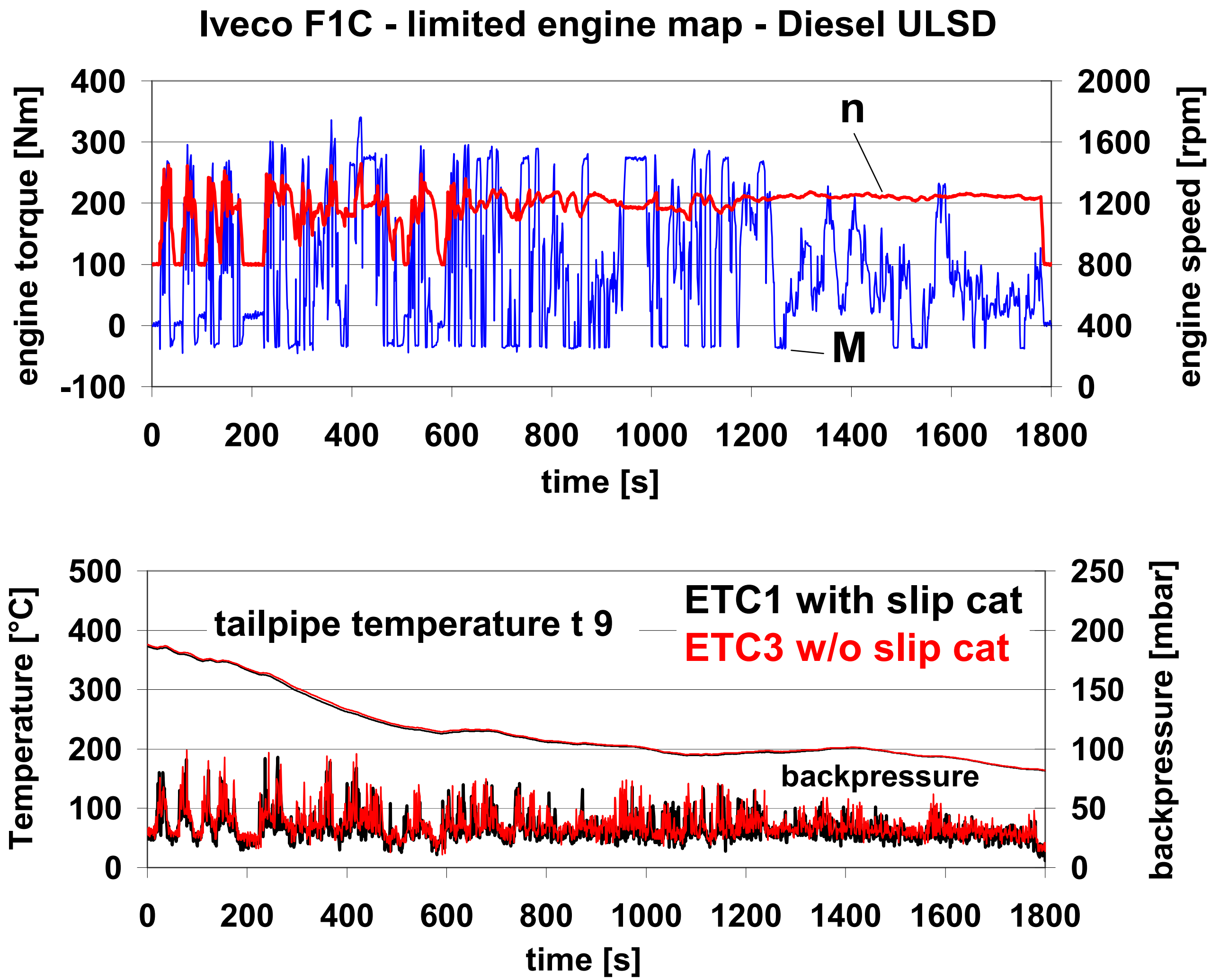
ETC 1 with slip with cat	ETC 3 w/o slip cat	ETC 4 Ref. w/o DPF + SCR

Conditioning before ETC

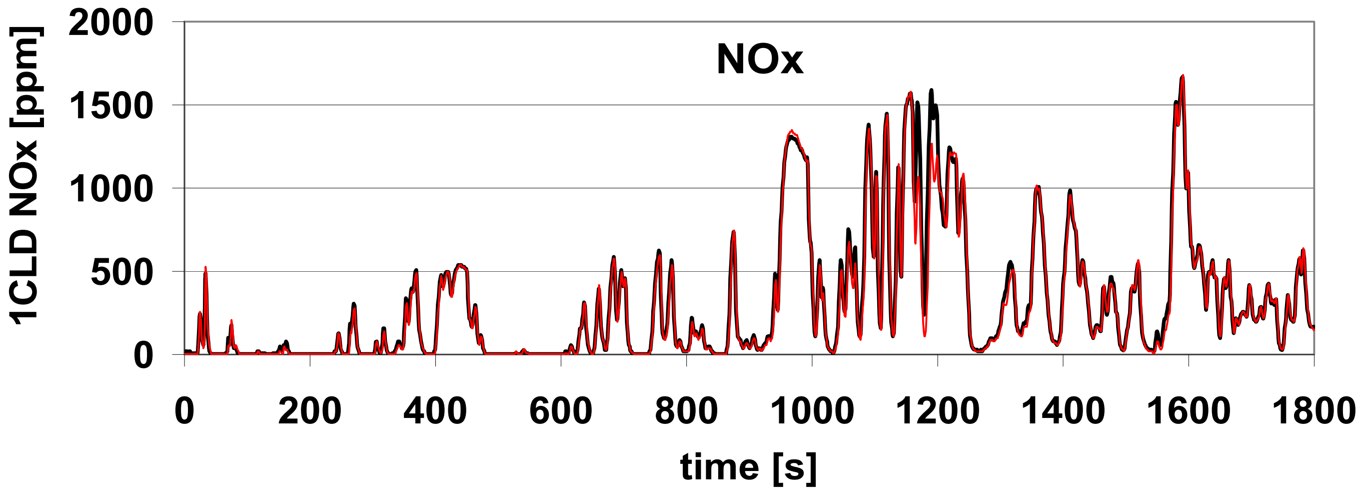
5 min → pt. 1 2200 rpm / 100 % Load

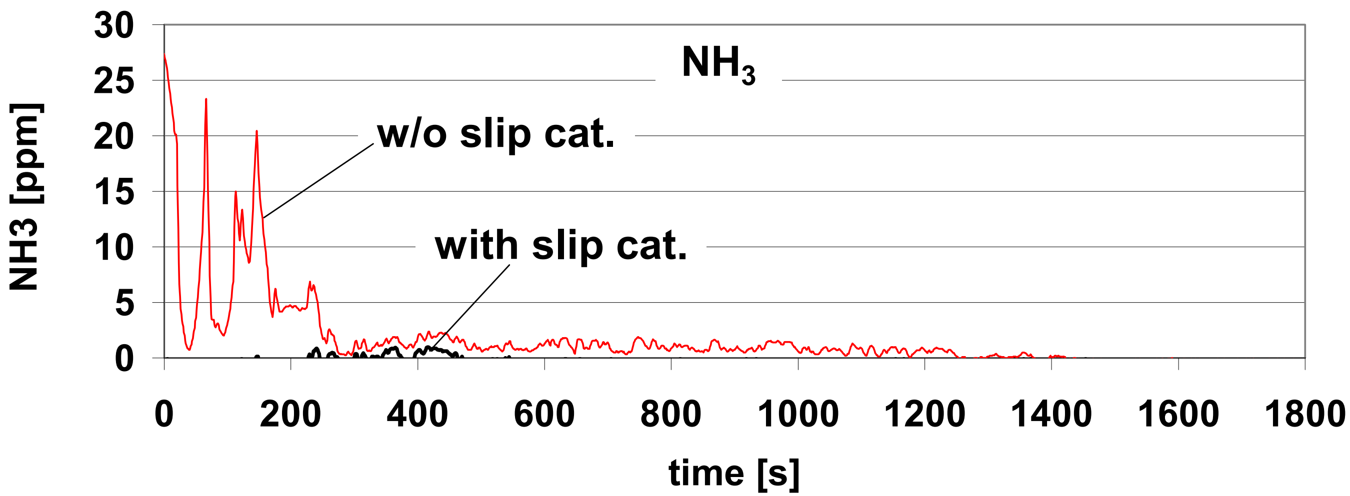
0.5 min → idle

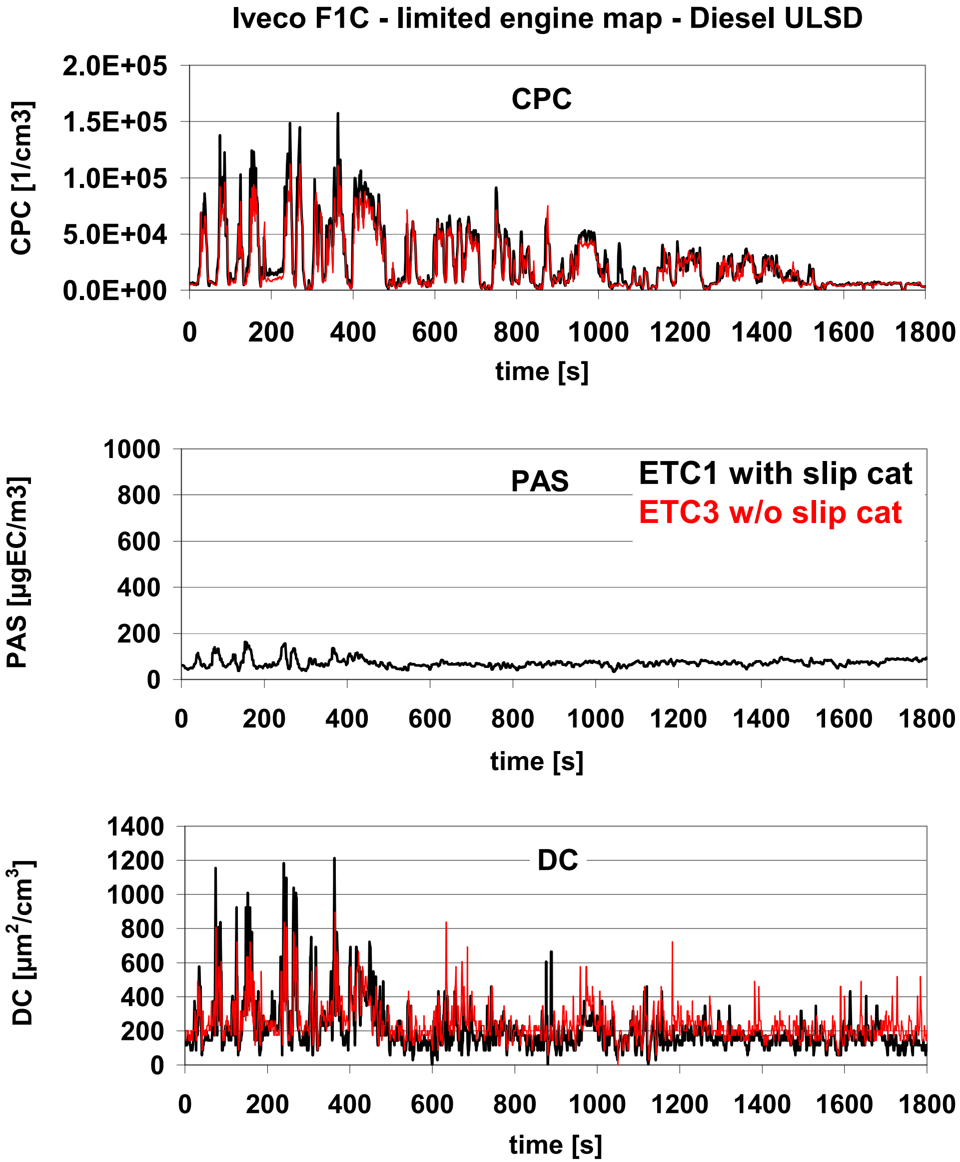
Comparison of 2 ETC's (ETC1-ETC3), with & w/o slip catalyst , α = 0.9



Comparison of 2 ETC's (ETC1-ETC3), with & w/o slip catalyst , α = 0.9







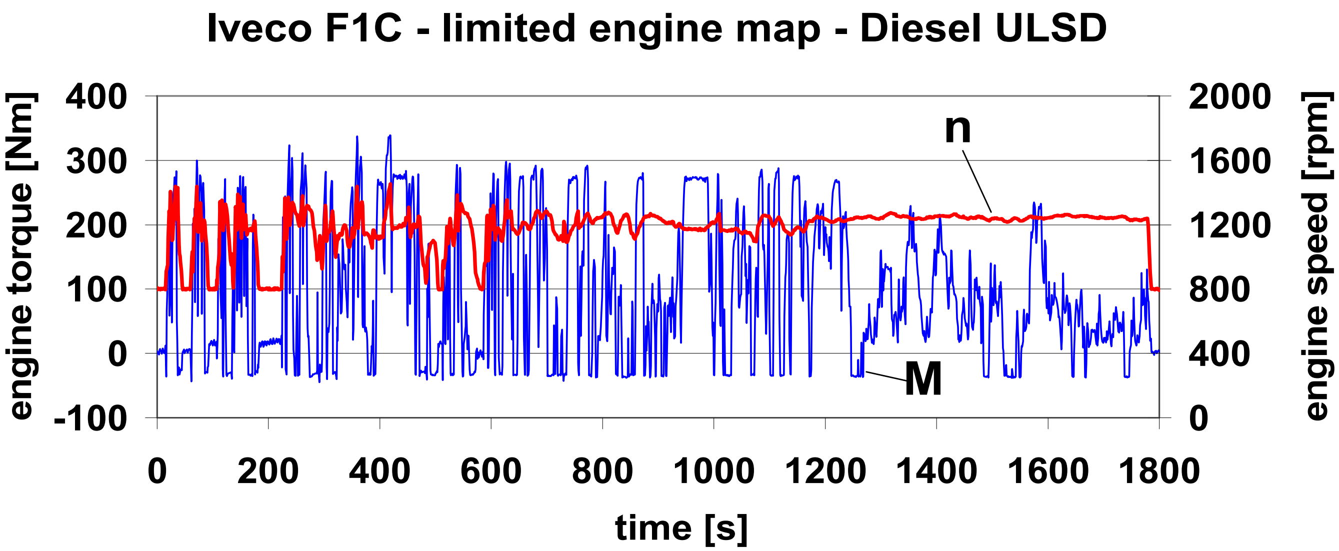
Comparison of

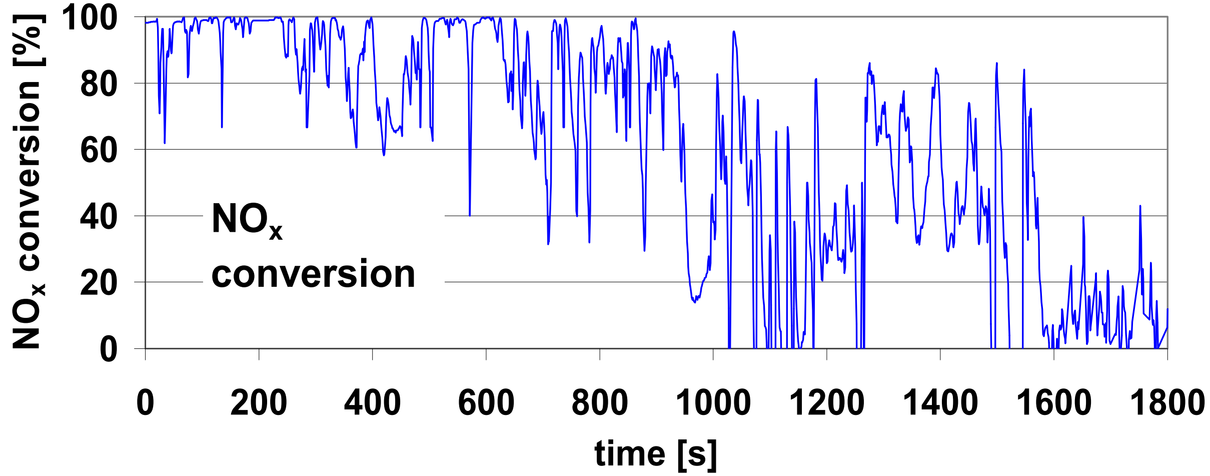
2ETC's (ETC1-ETC3), with & w/o

<u>slip catalyst</u>

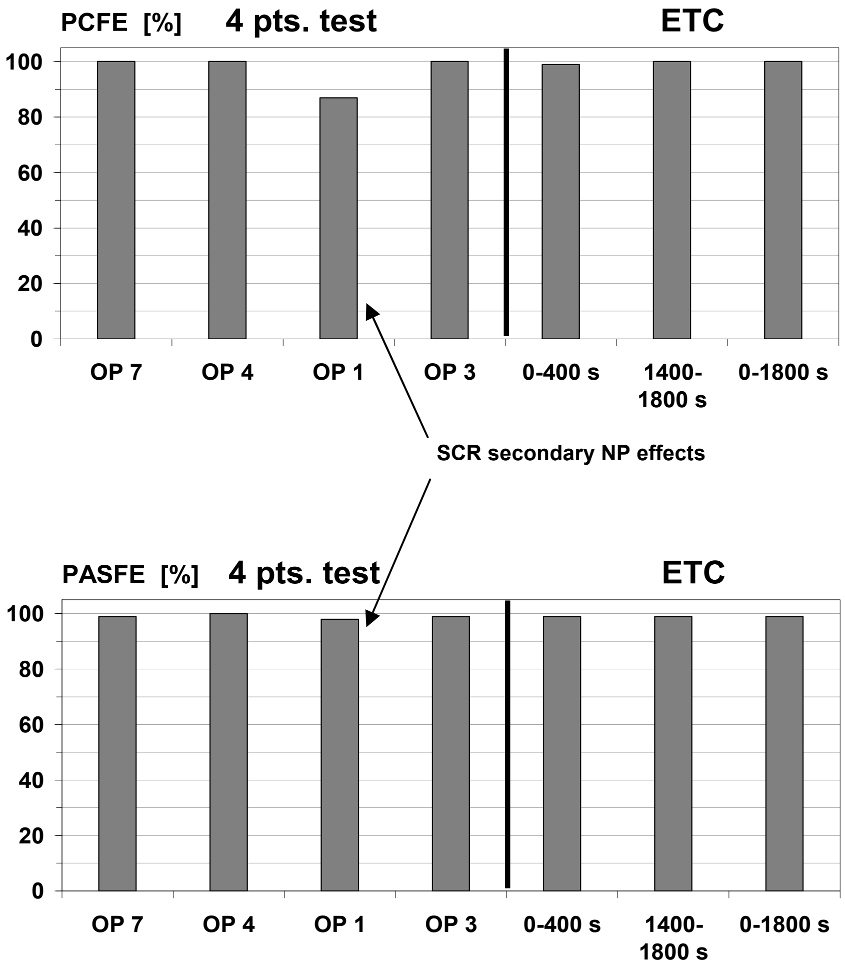
α=0.9

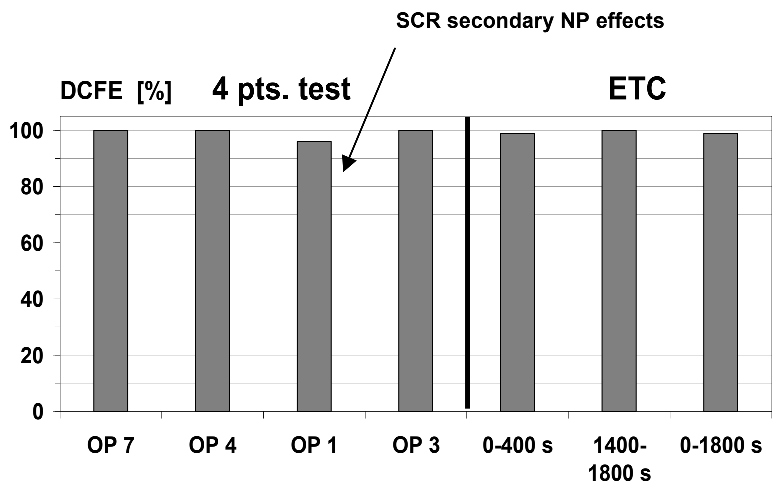
Comparison of 2 ETC's (ETC3-ETC4), reference & w/o slip catalyst , $\alpha = 0.9$





Filtration efficiencies of the combisystem after SCR – catalyst in stationary and dynamic engine operation





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IC-Engines and Exhaust Gas Control

Conclusion (1)

- $\alpha \uparrow \rightarrow$ **NOx** $\downarrow \rightarrow$ **NH3** $\uparrow$ **(w/o slip cat)**
- **urea switch on/off at lower t_{Exh}**
- **NO_x conversion rate in ETC dependent strongly on urea dosing = $f(t_{Exh})$**

further research and evaluations in course

Conclusion (2)

- **in ETC with α = 0.9**
  - average $NH_3 \leq 7$ ppm
  - average $N_2O \leq 3$ ppm
- **secondary NP**
- **DPF filtration efficiency up to100%**
  stationary = dynamic

further research and evaluations in course

SCR, V-HSO System

AVL

- Pre (Vor)-Catalyst
- Hydrolysis, SCR and Oxidation Catalyst

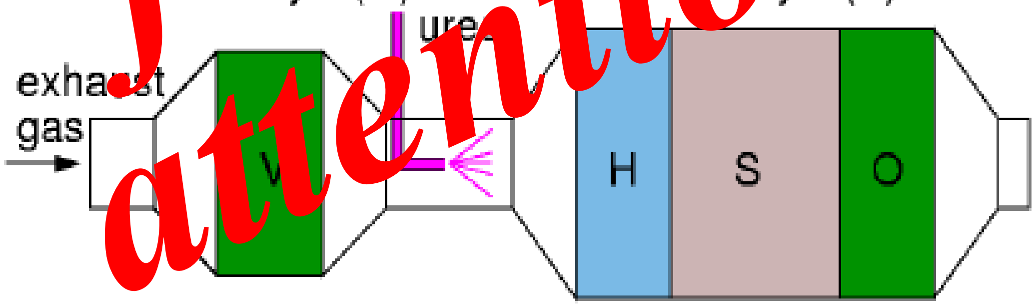
$$NH_3 + S \xrightarrow{k_6} NH_3(S)$$
$$NH_3(S) \xrightarrow{k_7} NH_3 + S$$

$$4NH_3 + 4NO + O_2 \xrightarrow{k_1,k_8} 4N_2 + 6H_2O$$ normal
$$4NH_3 + 2NO + 2NO_2 \xrightarrow{k_2} 4N_2 + 6H_2O$$ rapid
$$8NH_3 + 6NO_2 \xrightarrow{k_3} 7N_2 + 12H_2O$$ slow

$$(NH_2)_2CO(aq) \longrightarrow (NH_2)_2CO(s) + 7.1H_2O(g)$$

Thermolysis $$(NH_2)_2CO(s) \xrightarrow{k_{th}} NH_3 + HCNO$$

$$4NH_3 + 3O_2 \xrightarrow{k_4,k_9} 2N_2 + 6H_2O$$



$$2NO + O_2 \longleftrightarrow 2NO_2$$

$$HCNO + H_2O \xrightarrow{k_5} NH_3 + CO_2$$

$$4NH_3 + 3O_2 \xrightarrow{k_4,k_9} 2N_2 + 6H_2O$$

HNCO isocyanic acid

Thank you for your attention !