Conference report: 23rd ETH Conference on Combustion Generated Nanoparticles

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The 23rd ETH Conference on Combustion Generated Nanoparticles was held on June 17-20, 2019 in Zürich, Switzerland. The Conference program included about 45 presentations and keynote talks, as well as 88 posters on a number of topics, including particle fundamentals, engine emission control, particles from aircraft and other combustion sources, emission measurement, and health effects of air pollution. There were about 380 registered attendees. The conference included an exhibition with the participation of suppliers of emission control devices, emission measurement instruments, and related products and services.

The conference was officially opened by Prof. Gian-Luca Bona, the CEO of EMPA, Switzerland. His Opening Address—which briefly outlined the challenges faced by the world’s mobility sector—was followed by a Key Lecture by Prof. Wolfgang Lienemann of the University of Berne, titled *Environmental Ethics in the High Risk Society*, that aptly reflected the crisis in corporate ethics revealed by the Volkswagen emission scandal. After two days of technical sessions, on the last day of the conference, a Focus Event *Not just Diesel Soot → Detox all Combustion Engines* covered insights into toxic effects of all emissions from combustion engines, the importance of in-use emission control, as well as directions for future emission standards for new engines and vehicles.

Emission Measurement

The European particle number (PN) measurement procedure is being re-evaluated under the EU-sponsored DownTo10 (DTT) program [Andersson 2019]. The research aims to develop modified sampling and measurement methodology that would account for particles smaller than the current 23 nm cut-off point, for both laboratory (CVS) and RDE (PEMS) measurements. The political objective is to amend the EU regulatory limits for solid PN emissions to include all particles above 10 nm, down from the current 23 nm cut-off size. The DTT system is based on the existing PMP method, using either dilution + evaporation or a catalytic stripper + dilution to remove volatile particles from the sample. As before, a CPC is used for particle counting, and a 50% counting efficiency at 10 nm has been demonstrated. The program is now testing both lab-based instruments and portable exhaust particle sampling system (PEPS) prototypes. A number of measurements that have been performed show that most vehicles are compliant with the Euro 6 PN limit of 6×10^{11} 1/km for both > 23 nm and > 10 nm ranges. However, there are some technologies that can meet the current limit value in the PN_{23} range, but would fail in the PN_{10} range—in Figure 1, these points are located above the 100% PN_{10} line while below the 100% PN_{23} line. These technologies include mostly currently unregulated niche vehicle types such as performance motorcycles and CNG vehicles.

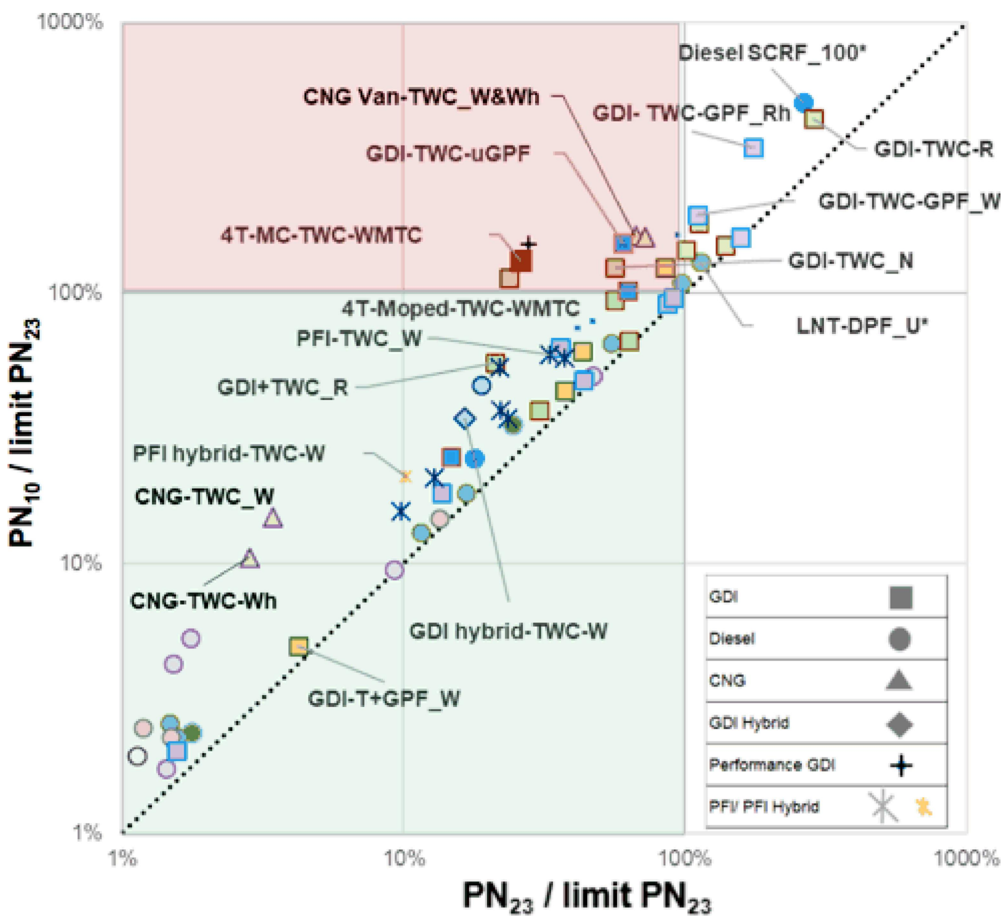

Figure 1. Comparative PN_{10} and PN_{23} data

Measurements were also conducted below 10 nm, with the instruments sensitive down to 4 nm. There were no excessive PN increases in the below-10 nm region in technologies with diesel or gasoline particulate filters (DPF or GPF), which shows that filters are effective in removing very small particles. On the other hand, high PN_{sub10} emissions were observed in several applications without particle filters—for CNG, over 130 times higher than the PN_{23} levels.

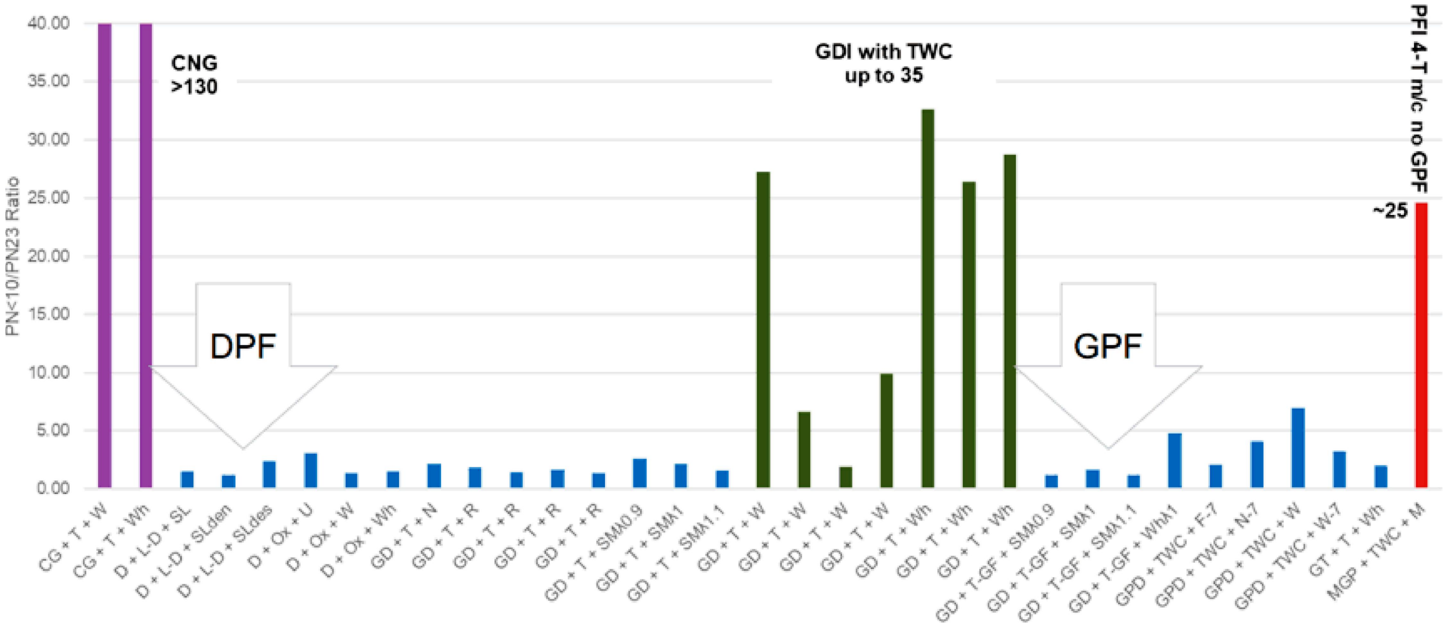

Figure 2. Ratio of particles below 10 nm to particles above 23 nm

All data where PN_{sub10} > 6×10^{11} 1/km

The updated methodology for the measurement of PN_{10} particle numbers was evaluated in a study conducted as part of another European sub-23 nm particle project, SUREAL-23 [Zinola 2019]. Experiments on a GDI engine showed that lowering the cut-off value from 23 to 10 nm was associated with an increase of PN emissions by about 10-20%. Interestingly, the PN increase was not observed downstream of a catalyzed GPF (cGPF), and the particle size distribution after the filter was shifted towards larger particles. It was hypothesized that the smallest particles, below 23 nm, were trapped and oxidized within the cGPF. The same study investigated the effect of hybrid drivetrains on PN emissions and found that PN emissions increased by a factor between 2.5 and 4. This increase was caused by the rapid high load transitions the thermal engine in hybrid operation.

Horiba discussed the development of two PN_{10} instruments—one for laboratory use and one for on-vehicle (PEMS) measurements—a development supported by the EU PEMS4Nano project. The devices utilize a catalytic stripper and a CPC. The laboratory PN_{10} system can be applied like the existing SPCS (PN_{23}) system, while two PEMS devices (10 nm and 23 nm) are still under evaluation; an ambitious target of ±20% for SPCS/PEMS correlation was achieved in some tests [Kreutziger 2019]. The diffusion charging (DC) sensor (Partector) that is at heart of two other PN PEMS systems—the AVL M.O.V.E. and NanoMet3 by Testo—has been also updated to the PN_{10} measurement range [Rüggeberg 2019].

The Southwest Research Institute (SwRI) has been evaluating a number of spark-plug sized exhaust sensors for particulate emission monitoring [Khalek 2019]. The work, conducted within the SwRI Particle Sensor and Durability Consortium (PSPD), supports the development of robust and accurate sensing technology and data transmission that could be used in future emission compliance mechanisms, and reduce the burden associated with current laboratory and in-use emission certification procedures. Three types of PM sensors are being evaluated: collecting, cumulative PM sensors based on resistance measurement (Bosch, Denso, Stoneridge); particle charging and net escaping current (NGK/NTK); and measurement of natural charge of particles (CoorsTek, formerly Emisense). Results were presented including sensors' steady-state and transient response, and sensitivity after multiple exposures. In future work, the SwRI plans to focus on NOx/NH_3 sensors, is a project with potential participation of the Truck and Engine Manufacturers Association (EMA), US EPA, and sensor manufacturers.

Two presentations were given on instruments for the measurement of ambient concentrations of carbonaceous aerosols. The FHNW University, Switzerland, has discussed the development of a new *total carbon* analyzer, dubbed FAst Thermal CArbon Totalizer (FATCAT), that collects particles on a filter in the form of a sintered metal tube. The collected sample is then heated, passed over an oxidation catalyst, and the carbon mass is determined via NDIR CO_2 detection [Keller 2019]. The Paul Scherrer Institute (PSI) reported on the next version of the Single Particle Soot Photometer - Extended Range (SP2-XR) for *black carbon* measurements. The updated instrument uses the same laser-induced incandescence (LII) principle as its predecessor, the SP2, but features a smaller size and weight, improved stability, extended detection range (50-800 nm), and real time automatic raw data processing [Bertò 2019].

Periodic Technical Inspections (PTI)

One session at the conference was devoted to the activities around the new periodic technical inspection (NPTI) initiative. In the EU, periodic technical inspections of in-use vehicles are mandatory, but the implementation and methods vary between countries. In general, emission checks during PTI rely on the vehicle's OBD information, which could be supplemented, for diesels, with a smoke opacity test. This existing approach, however, is not effective in identifying emissions tampering—such as removing the DPF or disabling the SCR system—or detecting failed aftertreatment components. As a result, several countries are considering a mandatory emission test at the PTI, designed to identify such high emitting vehicles. In 2016, through an initiative of the VERT Association, an informal NPTI working group was established to develop suitable test procedures. For DPF testing, this is envisioned as a particle number test performed at the tailpipe at low idle with a PN emission limit in the range of $1 - 5\times10^5$ cm^{-3}, which correlates with Euro 6 type approval data.

The Netherlands will likely become the first EU country to introduce NPTI testing requirements for PN emissions [Kadijk 2019]. The Dutch PTI protocol, expected to be officially published in September 2019, includes a PN limit of 250,000 cm^{-3}, to be met at low idle. The specifications for PN testers developed by the Dutch metrology authority NMi specify three particle sizes (23, 50, and 80 nm) and a concentration range of 0 - 500,000 cm^{-3}. The PTI PN test will be implemented in the Dutch PTI as soon as sufficient quantities of new PN testers are available on the market, which is expected for 2021. Other countries expected to adopt NPTI emission tests include Germany and Switzerland.

TNO also presented an interesting study of in-use emissions from older gasoline vehicles [Kadijk 2018]. A large percentage of tested vehicles emitted elevated, in some cases significantly elevated, levels of NOx, Figure 3. For comparison, the respective Euro 3/4/5 NOx certification limits are 0.15/0.08/0.06 g/km (Euro 2 has a NOx+HC limit of 0.5 g/km). The reason for the increased NOx was not clear, but high emissions occurred more often in lean burning cars, and might have been linked to a faulty oxygen sensor.

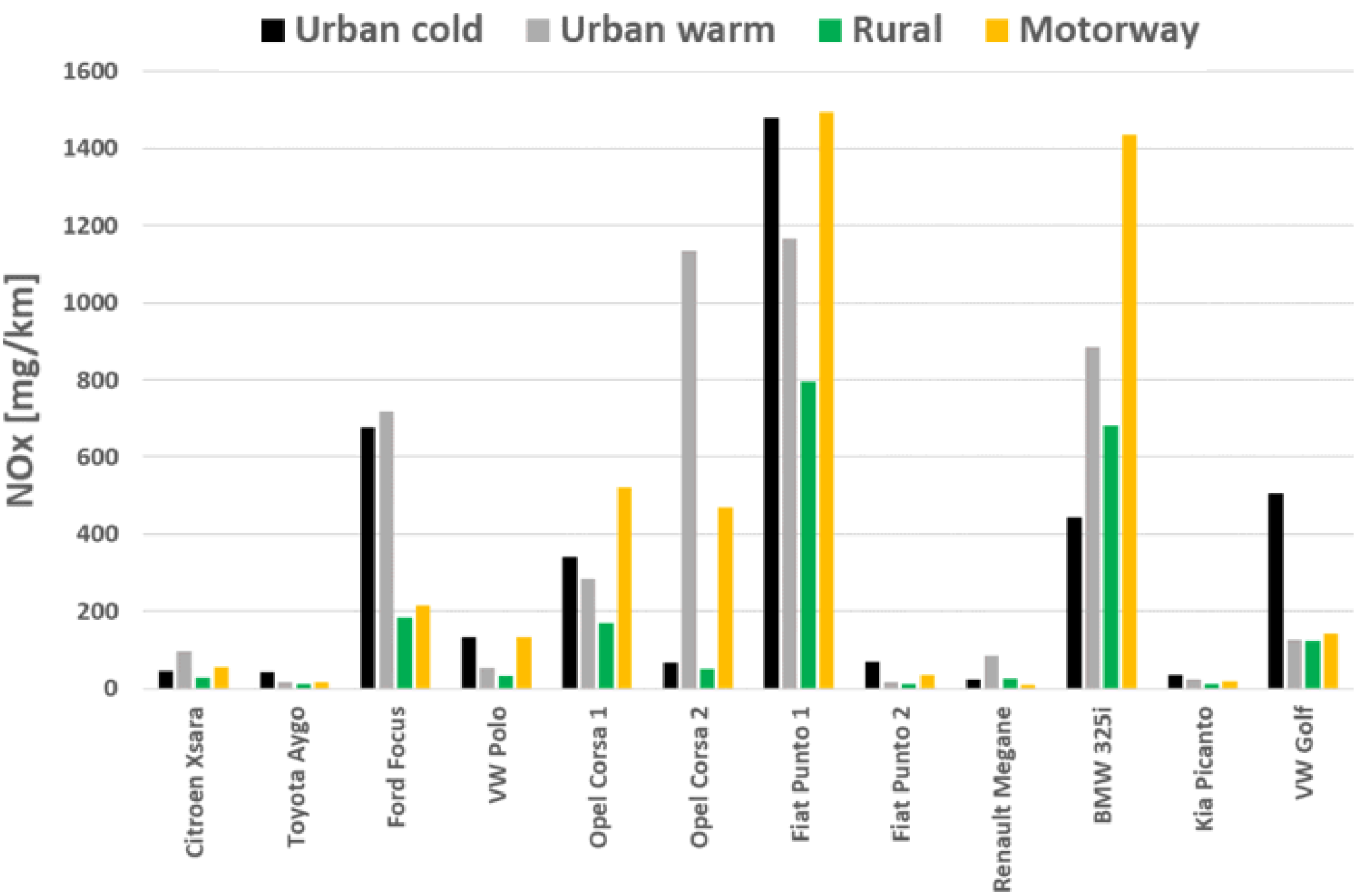

Figure 3. NOx emissions from old gasoline vehicles

Common Artemis Driving Cycle; 12 Euro 2-5 vehicles; Mileage: 155,000 - 254,000 km; Age: 6 - 20 years

The PTI test should be quick and cost-effective, which means the use of a chassis dynamometer should be avoided. While such a quick and accurate test is possible for PN emissions—and suitable, cost-effective PN instruments are beginning to be available—a PTI test for diesel SCR systems remains problematic [Mayer 2019]. Options that have been considered to warm up the exhaust system to enable urea injection include a load step test [Czerwinski 2019], using thermal management tools at idle (intake or exhaust throttle, late fuel injection), or driving the car at high speed on the road immediately before the test.

Aircraft Emissions

The United Nations' aviation agency, ICAO (International Civil Aviation Organization), is introducing the first-ever global regulatory limits for aircraft engine particle mass and number emissions [Rindlisbacher 2019]. From 1st January 2020, all in-production engines (larger than small business jet size) will need to be certified for emissions of *non-volatile particle mass* (nvPM) and *nonvolatile particle number* above 10 nm (nvPN)—with no limits yet. Regulatory limits for both emissions were adopted in February 2019 and are to become mandatory from 2023. Nonvolatile particles are defined as those that do not volatilize when heated to 350°C. Emissions are determined over a standard landing and take-off (LTO) cycle, with four engine modes: take-off (100% thrust, 42 s), climb (85% thrust, 132 s), approach (30% thrust, 240 s), and taxi (7% thrust, 1560 s). The standards bring special challenges for combustion technology, as the nvPM value can have different trends in different engines. Lean burn combustion technology can address both NOx (which has been regulated for years) and nvPM, but technologies are not scalable to the full range of engine sizes.

Due to the extreme conditions during aircraft engine testing, the sampling lines must be very long, which contributes to severe sampling losses—up to 80-90% sample loss at 10 nm [Kittelson 2019]. Therefore, the line loss correction is of critical importance. The correction method requires the knowledge of effective particle density, which is measured in a set-up including a DMA (differential mobility analyzer), CPMA (centrifugal particle mass analyzer), and a CPC (condensation particle counter). Two configurations are possible: DMA-CPMA-CPC (more common) or CPMA-DMA-CPC (faster mass scan). However, particle density results obtained in these two configurations for a similar engine and fuel conditions were notably different, and the reasons for the differences remain elusive.

A study at the Tokyo Narita airport investigated the composition of aircraft engine nanoparticles [Fushimi 2019]. Samples were collected near a runway during the day, in the presence of air traffic, and at night, when there is no air traffic at Narita. The composition of the day samples, as determined by mass chromatography, was similar to the hydrocarbon makeup of common jet lubrication oils. It was concluded that aircraft nanoparticles were composed mostly of organic carbon (OC), and that about 50% of the OC was derived from the jet lubrication oil.

Health Effects

Prof. Nino Künzli of the Swiss Tropical and Public Health Institute (Swiss TPH) has called for the adoption of strong clean air policies and global air quality standards based on the WHO exposure recommendations [Künzli 2019]. The EU clean air policy has been described as "semi-ambitious"—the EU promoted (dirty) diesel until recently, delayed the introduction of DPF-forcing emission standards, and showed insufficient control of the industry, as demonstrated by the Volkswagen emission scandal. A central failure of EU clean air policy are the European exposure limits for PM_{10} and $PM_{2.5}$—values not informed by science, but by the vested interest of the auto and coal industries. These EU limit values are twice as high as the WHO guideline and the respective PM limits in many other countries: for PM_{10}, the EU limit is 40 μg/m^3 (annual mean), compared to 20 μg/m^3 WHO value; for $PM_{2.5}$, the respective figures are 25 μg/m^3 and 10 μg/m^3. Prof. Künzli was very critical of many policies adopted in the aftermath of the VW scandal. The low emission zones ("diesel bans") adopted in some cities were strategically located to protect air monitoring stations from non-compliance, rather than to protect public health. Several of the "derailed" emergency policies require upgrading to newer cars, which benefits the auto industry, while violating the principles of social justice, as the poor have no means to pay for new vehicles. As a result, public unrest erupted in some German cities, with groups—supported by some "manipulated" German media—calling for relaxing air quality limits. On a global scale, German "bans" result in exports of older cars to Eastern Europe and Africa. Such *outsourcing of pollution* is one of the reasons behind the ongoing deterioration of air quality in many poorer nations, and should not be an acceptable policy for the West.

Commenting on the health effects of ultrafine particles (UFP, particles < 100 nm), Prof. Künzli emphasized that claims that UFPs are more harmful than other pollution components are counterproductive, because the ultimate objective is to ensure that the air is clean of all pollutants—PM_{10}, $PM_{2.5}$, UFPs or NO_2. This topic was further discussed in the following presentation, which reviewed the existing body of literature on the health effects of UFPs [Probst-Hensch 2019]. There is "suggestive evidence" for independent *short-term* health effects of UFPs on inflammation, autonomic tone and blood pressure. On the other hand, there is insufficient evidence on short-term effects on mortality, and insufficient evidence to draw firm conclusions on *long-term* effects of UFP—beyond those from $PM_{2.5}$ exposures. A new study on UFPs conducted in Ontario, Canada [Weichenthal 2019] found some health effects (mostly cardiovascular) other than those from exposure to $PM_{2.5}$ or NO_2, as well as new evidence suggesting a possible link between UFPs and brain cancer.

Engine Research

A new generation of gasoline DI engines is being developed by the European PaREGEn consortium, with a target to reduce CO_2 emissions by 15%. Two demonstration vehicles, one Mercedes E180 and one Jaguar XE, will comply with Euro 6 RDE limits with PN emissions measured to a 10 nm size threshold. As part of the project, a team from ETH Zürich / Vir2sense has been working on a virtual gasoline particle sensor (vGPS) for direct injection SI engines [Barro 2019]. The virtual sensor estimates PN/PM emissions based on input parameters from the ECU and actual sensors (such as intake p & T) utilizing models for spray formation, wall film evaporation (impingement), combustion, and soot formation. The vGPS has been validated on a single cylinder naturally aspirated engine. Future plans include calibration on a multi-cylinder turbocharged engine.

Other noteworthy engine research work included an investigation of diesel-natural gas RCCI combustion, where it was shown that increasing the port fuel energy portion (i.e., NG) reduced PN emissions [Hosseini 2019]; and a study of an IC engine operated with a hydrogen-rich gaseous fuel (direct or port injection) obtained via steam reforming of methanol using exhaust gas heat to increase efficiency [Thawko 2019].

Aftertreatment

The European Commission Horizon Prizes program includes a *Cleanest Engine Prize: Design for the Future* (a *safety net* alternative to electrification if growth is slow—apply by 20 August 2019) and a *Retrofit Prize* to clean high emitting vehicles on the road [Maggiore 2019]. The latter prize has been awarded last year for a Euro 5 vehicle retrofit with Amminex NH_3 storage and delivery system and Johnson Matthey SCR catalyst. Potential future retrofit areas envisioned in the presentation include GPF retrofits of gasoline vehicles, which can produce particles that are smaller than diesel and contain more PAHs, and even natural gas engines, with large PN shares escaping counting due to small size (3-10 nm) yet total count similar to unfiltered diesel.

The Danish Environmental Protection Agency (EPA) has started a project to reduce emissions from ships in Danish waters. One ship—the inland ferry M/F Isefjord—has been already fitted with Exilator DPFs and switched to a 50 ppm sulfur MGO fuel [Køcks 2019]. The DPFs on main engines burn soot in normal operation (passive), while DPFs on generators burn soot at periodically increased load (controlled automatically). The expected ash cleaning interval is once a year. Both types of DPFs were found to reduce PM and PM emissions by over 99%, and to effectively reduce exhaust noise. The next demonstration will involve the installation of Purefi SCR and DPF systems on World Mistral (with 1,000 ppm S fuel, which presents a challenge for the selection of DPF technology), followed by another DPF+SCR installation on M/S Pernille, with 50 ppm S MGO fuel.

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The next, 24th ETH Conference on Combustion Generated Nanoparticles is scheduled for June 22-25, 2020 at ETH Zürich, Switzerland.

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