Ultra-Low PN_{10} Emissions of a Close-Coupled Emission Control System on a Heavy-duty Truck Application

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Heavy-duty vehicles represent a significant portion of road transport and they need to operate in a clean and efficient manner. Their exhaust emission control systems need to be enhanced to sustain the high conversion efficiencies seen during motorway conditions to other operating conditions.

The European Commission is developing legislative proposals for Euro 7 and Euro VII emissions regulations for light- and heavy-duty vehicles. The new Euro VII regulation will likely focus on ensuring the emissions from heavy-duty vehicles are minimized over extensive on-road operating conditions and in particular on operating conditions such as urban driving and cold start. These challenges are increased by the need to ensure low secondary emissions like PN_{10}, NH_3 and N_2O as well as low impact on CO_2 emissions.

The low pollutant emissions achieved by an AECC heavy-duty Diesel demonstrator vehicle will be presented. The vehicle is equipped with an innovative layout of state-of-the-art emission control technologies, combined with an advanced engine strategy implemented to an existing Euro VI-C long-haul truck. The new emissions control system integrates a close-coupled Diesel Oxidation Catalyst (DOC), a first Selective Catalytic system Reduction (SCR) in close-coupled position, a 2^{nd} DOC followed by a catalyzed Diesel Particulate Filter (DPF), and the 2^{nd} SCR from the dual-SCR system with twin AdBlue® dosing controlled by FEV developed software. Both SCR systems contain an Ammonia Slip Catalyst (ASC).

The presentation will focus on the sub-23 particles emissions PN_{10} measured over a broad range of operating conditions, including cold start and urban driving as well as for different payloads and ambient temperatures.

To show the potential CO_2 reduction on life cycle assessment basis, the vehicle has also been tested with a 100% renewable fuel. This, in combination with engine efficiency improvements, should enable the next generation heavy-duty vehicles to operate with ultra-low pollutant emissions, whilst maintaining their path towards the required CO_2 targets.

Particle emission from direct injection internal combustion engine fed with various gaseous fuels

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Introduction & Background: Gaseous fuels as hydrogen and methane are considered most frequently as possible alternatives to conventional liquid fuels. Besides, on-board production of a hydrogen-rich reformate using Thermo-Chemical Recuperation improves thermal efficiency and reduces emissions of gaseous pollutants. Commonly, gaseous fuels are introduced into the ICE intake manifold through a port fuel injection (PFI). The latter results in improved combustion and concomitantly reduced exhaust emissions, as compared to liquid fuels. However, PFI may lead to abnormal combustion phenomena, as well as maximal power loss. Direct injection (DI) of gaseous fuel eliminates these problems, and contrary to the PFI method it reduces the compression work caused by the additional volume of gaseous fuel in the intake manifold. Most of previous studies dealt with particle emissions of IC engines fed with gaseous fuels, considered fuel supply to the engine intake manifold. These works reported on the reduction of particle emission as compared to gasoline. There is a limited number of studies investigating particle formation in DI engines fed with gaseous fuels, and no comparative information is available on the effect of fuel type. The main goal of this work is to investigate particle formation in a direct injection SI engine fed with different gaseous fuels like hydrogen, methane, and hydrogen-rich reformate (containing 75% mol. hydrogen and 25% mol. CO_2). The effect of the end of injection timing (EOI) and fuel type was investigated, and a comparison to PFI with the same fuels was performed.

Methodology: The experimental setup was based on a single-cylinder Lister-Petter AD1 4-stroke engine with a high compression ratio (r=15.5) modified for spark-ignition operation. Particle number concentration and size distribution measurements were performed with TSI model 3090 Engine Exhaust Particle Sizer (EEPS) Spectrometer. A direct experimental comparison was performed between the PFI and DI fuel supply methods and different gaseous fuels: hydrogen, methane, and the reformate.

Results & Conclusions: The results showed that the total particle number emitted from the engine fed by DI of the hydrogen-rich reformate was the highest among the studied fuels. Besides, hydrogen fuel, with no carbon content, resulted in a higher total particle number compared to methane. The increase in particle emissions was noticed especially at high engine loads, while for the lower engine loads, which can be achieved using both PFI and DI methods, no significant difference was noticed between the various fuels. The direct experimental comparison of particle emission between the reformate port and direct injection on the same engine elucidates that the increase in particle formation is a result of the applied direct injection method. Notably, the reformate fuel requiring the longest injection duration due to the lowest fuel heating value resulted in the highest particle emission. Studying the effect of EOI with the DI method for the various fuels revealed the same trend of excessive particle formation at high engine loads. The retarded EOI timing (-70 CA BTDC) with longer injection duration resulted in a higher particle emission compared to the earlier EOI timing (-100 CA BTDC). The observed higher particle formation with direct reformate injection is attributed to the lubricant involvement in the combustion process. The fuel jet developed during the injection causes lubricant vapor entrainment into the jet, while the jet impingement on cylinder walls causes oil removal from the walls. This lubricant is mixed in the cylinder bulk which led to incomplete combustion with resulting excessive particle formation.

High Dimensional Fast-Response Particle Number (PN) Surrogate Model Building Methodology for Heavy Duty (HD) Diesel Engine Applications

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India's existing BS-VI emissions standards on sub-23 nm particle number (PN) emission limit applies to the engine testbed level. As heavy-duty (HD) diesel engines are always fitted with Diesel Particulate Filter (DPF), meeting PN emissions legislative limits at the tailpipe level is not difficult. However, future legislation requirements may be similar to European on-road In-service Conformity (ISC), by using Portable Emission Measurement System (PEMS) for vehicle level PN measurements. Therefore, it is an important aspect to understand the physics involved in the generation and evolution of particles, the corresponding sensitive parameters, and therefore to develop a capability to robustly estimate engine-out PN for HD diesel engines. This technical work is focused on estimating the engine-out PN for HD diesel engine by using the MoDS-SRM Engine Suite digital workflow. The purpose is to build detailed physico-chemical engine models and a high dimensional, fast-response surrogate model that is capable of accurately capturing combustion characteristics and engine-out emissions at steady-state operating points. The PN measurement data is collected with AVL APC 489 device, covering various operating conditions with distinct engine speed and load across the entire operating window of a HD compression ignition diesel engine. The physico-chemical models are calibrated (parameter estimation) as part of the MoDS-SRM Engine Suite workflow with 40% of the measured operating points. Subsequently, the calibrated detailed models undergo validation (blind-testing) against the remaining 60% of the measured operating points. The calibrated models are able to simulate the validation points with satisfactory accuracy, capturing the general trend across various operating conditions for gas-phase and particulate emissions. For particulate emissions, 80% and 62% of the validation points are within 50% error in relation to the measurement data for soot mass and PN. Had it been a purely statistical or data-driven model, approximately 85%-95% of the measured operating points would be required for model calibration to reach the level of accuracy that is achieved in this work. In order to reduce computational expense further, a fast-response model is developed to simulate steady-state and transient operating conditions. The surrogate model generated from the MoDS-SRM Engine Suite digital workflow is applied to perform transient cycle simulations in MATLAB. The future scope of the work includes the addition of more measurement data with respect to variations in EGR, injection pressures, high and low ambient temperatures, high altitude, and cold zones to demonstrate the cost-reduction potential of the digital workflow in some of the aforementioned measurements-intensive scenarios.

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Applying lessons learned from diesel exhaust to brake wear nanoparticle measurements and regulation

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Automobile friction brakes generate, in addition to coarse particles generated by mechanical processes, highly variable amount of nanoparticles from high temperature processes. In this work, four sets of front brake pads for a typical mid-size passenger car were subjected to selected parts of standardized brake performance tests believed to be reasonably realistic for common driving, and one set of pads also to the newly developed WLTP brake wear testing procedure. Tests were done on a brake dynamometer with an enclosed chamber and particles were measured in the duct serving at the outlet of the cooling air. A fast electric mobility particle sizer, checked to provide a reasonable response to different types of non-carbonaceous nanoparticles, was used to measure particle size distributions. The particle production was found to vary over about 5 orders of magnitude on both per-stop and per-kWh basis, with higher intensity, higher total energy dissipated, and higher temperature all correlated with a non-linear increase in emissions. While the emissions over the WLTP cycle were relatively low, the very high contribution of more aggressive, yet still realistic braking events, even though relatively infrequent, should not be overlooked. Just like with driving a non-DPF vehicle, aggressive, high-speed driving produces substantially more particles than defensive, gentle, do-not-spill-your-coffee driving.

Solid and volatile brake-wear nanoparticles under real-world operating conditions

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Recent studies have identified brake-wear as the major contributor to road transport Particulate Matter (PM) emissions, as a result of an efficient implementation of exhaust PM regulation worldwide. Accordingly, there has been a growing interest in the characterization of brake-wear emissions over the last years. In an attempt to establish a standardized test procedure that would facilitate comparison of results from different studies, the United Nations Particle Measurement Programme Informal Working Group (PMP IWG) has been actively working on the development of a harmonized brake-dynamometer methodology, including a novel test cycle (WLTP-Brake).

In this study we have applied the proposed PMP IWG methodology for the characterization of particulate emissions of the same brake system on two different dilution tunnels. The two tunnels were designed to operate at different flowrates (170 to 270 m^3/h compared to 300 to 1800 m^3/h). The brake system was tested under the WLTP-Brake cycle but also under another real-world brake cycle derived within the Lowbrasys European research project from analysis of the Los Angeles City Traffic (3h-LACT). The effect of bending procedure on particle emissions was also evaluated through dedicated tests in which the last and more demanding section of the WLTP-Brake was employed for the conditioning of the brake.

Particulate measurements included $PM_{2.5}$, PM_{10} as well as particle number (PN) following the recently introduced exhaust PN methodology that extended the detector cut-off size to 10 nm. Measurements also included real time size distributions and number concentrations of thermally untreated samples. Disc temperature profiles were investigated with a thermographic camera and an array of thermocouples embedded on the brake disc.

Both PM_{10} and $PM_{2.5}$ emissions were found to agree within 20% at the two facilities, despite the vastly different operating flows of the tunnels. Similarly, no statistically significant effect of the cycle could be identified on the PM emissions despite the nearly two times higher average disc temperatures over the LACT cycle (~110°C compared to 60°C over the WLTP). Most of the airborne PM was larger than 2.5 µm, with the ratio of PM10 to PM2.5 raging between 3.5 and 4.

Elevated concentrations of nanosized particles, thermally stable at a catalyst operating at 350°C, were released over the WLTP-Brake, under specific burnishing procedure. Their nature and formation pathways are currently unclear. Volatile nanoparticles were observed over the 3h-LACT cycle but only at the high tunnel flow. Their relative concentration decreased with increasing tunnel flow. This behavior is in accordance to nucleation theory since increased tunnel flows imply increased dilution of vapor precursors and potentially reduced release of vapors through more efficient cooling of the brake disc.

The development and convergence of co-pathologies associated with Tau, Aβ, α-synuclein and TDP-43 proteinopathies in Metropolitan Mexico City children and young adults: a health crisis is in progress. Nanoparticles a common denominator?

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Complex interacting pathways play key roles in Alzheimer's disease and other fatal neurodegenerative diseases, wherein we have multiple misfolded proteins causing a range of neuropathological changes and contributing strongly to clinical symptoms, including cognition deficits as well as brain MRI, gait and equilibrium, olfaction and brainstem auditory evoked potentials (BAEPs) alterations.

Quadruple misfolded proteins (tau pre-tangles and neurofibrillary tangles, amyloid-β [Aβ], α-synuclein, and transactive response DNA-binding protein 43 [TDP-43]) in the same brain are common in children and young residents in Metropolitan Mexico City (MMC) exposed to high concentrations of fine particulate matter $PM_{2.5}$ and nanoparticles. Indeed, 99.5% of 203 consecutive forensic autopsies in subjects younger than 40y, exhibit AD hallmarks, 20% Parkinson's disease and 18.7% TDP-43 pathology. Cortical tau pre-tangles, neurofibrillary tangles (NFT) Stages I-II, and amyloid phases 1-2 are documented by the 2sd decade. Of critical importance is the documentation of NFT stages III-V in 24.8% in 30-40 y old subjects.

Cognitive changes in subjects age 21.6±5.8 years are likely an indication of the neuropathology seen in forensic young cases. The Montreal Cognitive Assessment (MoCA) administered to 517 urbanites, showed an overall MoCA score of 23.92±2.82 (normal 26-30), with 24.7% and 30.3% individuals scoring ≤24 and ≤22, respectively (Mild Cognitive Impairment MCI≤24, Dementia scores D≤22). Cognitive deficits progressively targeted Visuospatial, Executive, Language, and Memory domains.

Alzheimer Continuum subjects have higher numbers of brain NPs versus clean air controls with normal brains. Iron rich NPs and transitional metals and non-metals are identified in neural cells and endothelium's mitochondria, Golgi, and endoplasmic reticulum and are associated with significant structural organelle damage.

We strongly support combustion and industrial NPs are key players in early ROS generation, neurovascular unit, mitochondria, endoplasmic reticulum and endolysosomal dysfunction, and catalysts for protein misfolding, aggregation and fibrillation. Fe-rich NPs respond to external magnetic fields and thus might be involved in cellular damage by agglomeration/clustering, magnetic rotation and/or hyperthermia.

Nanoparticle exposure regardless of sources carries a high risk for the developing brain homeostasis and ought to be included in the AD, PD and TDP-43 research framework. The ultimate neural damage and neuropathology could depend on NP characteristics and the differential access and targets achieved via their portals of entry. Control of NP sources becomes critical.

Neurodegenerative fatal diseases are likely the result of complex interactions between environmental and genetic factors. A more complete understanding of NPs as plausible and modifiable environmental risk factors for the development of diseases such as Alzheimer and Parkinson's, Frontal-Temporal Dementia and Amyotrophic Lateral Sclerosis evolving from childhood will help guide their early detection and prevention. We are in the midst of a devastating crisis too big to ignore, with profound health, social and economic consequences.

Differentiating Translocated Exogenous vs Bio-generated Endogenous Nanoparticle Types in Olfactory Bulb of Humans with Neurodegeneration

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An increased risk for Alzheimer's and related dementia has been hypothesized to be causally associated with ambient PM air pollution. Aerosol constituents in polluted air contain redox-active metal- and other nanoparticles (NPs), and mounting evidence supports their role via neuronal nose to brain transport for neurodegeneration at and beyond the rostral (olfactory bulb (OB)) and caudal (brain stem) point of CNS entry. We examined autopsied OBs to determine whether inhaled pollution-derived NPs reach the brain via the olfactory nerve. To understand the accumulation of ambient nanoparticles in the OB requires to differentiate between the sources that are coming either from exogenous (aerosol pollution) or endogenous origins (biomineralization). An example are iron NPs for quantifying the various iron components which is crucial to link exogenous iron to neurodegeneration. Our approach uses analytical high-resolution transmission TEM of OB thin-sections coupled with electron energy loss spectroscopy (EELS) to assess compositions, relative quantities, and redox-activities of distinct NPs and to identify the physiochemical fingerprints of individual iron NPs that either translocated to OB regions (exogenous) or precipitated in vivo (endogenous). We also analyzed particles in OBs that seem to have originated from specific occupational aerosol exposures, for example tungsten particles that were present predominantly in sub-nano scale (0.3-1.5 nm) inside glomeruli and axons. Analyses were performed using a cryo-stage to stabilize the biological sections. We grouped NPs with identical characteristics and used digital imaging and spatial statistics to assess their frequency in the OB. Information on exogenous NPs locations and interactions with axons, mitochondria, amyloid plaques and or Lewy bodies were systematically correlated with Subjects that have well-documented impaired olfaction and dementia. Exogenous silica NPs are typically associated with heavy metal inclusions (Fe, Mn, Zn, Co, Ti, Zr, Cr, As, Se, Pb, among others) which typically occur as ultrafine dispersions and co-translocated (Trojan Horse Mechanism) to OB of select subjects. Exogenous iron NPs included iron oxides and phosphates. The location and frequency of endogenous iron NPs in OB (iron oxyhydroxide; ferritin) are related to the occurrence of amorphous silica, alumina, and carbon NPs as well as heavy metal NPs and may be a marker for inflammation and oxidative stress.

The Impact of Ultrafine Particles on Mental Health

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While there is robust evidence of the critical role of $PM_{2.5}$ in mental health and psychiatric disorders, much less is known about the specific role of ultrafine particle exposure. Lack of assessment of ultrafine particles (UFP) makes it difficult to attribute adverse effects specifically to this size range of particulate matter. This is despite a wealth of predominantly short term epidemiological studies indicating that some of the adverse effects of particulate matter on health are strongly mediated by UFP. The objective of this review is to summarize evidence of the role of UFP in cognitive and mental health from studies of humans and related animal models. Strategies to enhance both human and animal studies through increased collaborative interaction are also discussed. Human studies of UFP clearly indicate the potential for adverse effects on mental health. In areas where $PM_{2.5}$ concentration exceeds government standards, there are increased risks of impaired cognitive development, autism, Alzheimer's/dementia, depression, and ADHD.

Air pollution and UFP specifically may cause cognitive deficits in both early and late life. Compared to other research areas where there are often differences between the findings of clinical and animal studies, work in animal models of the effects of UFP on mental health have been consistent with, and supportive of, the epidemiological and clinical findings. The further optimization of lab related experimental studies through the expansion of naturalistic paradigms and increased direct collaboration between basic researchers and clinicians will enhance the collection of translationally relevant data. Taken together, these human and animal studies indicate that elevated concentrations of UFP air pollution have a considerable adverse impact on the brain and mental health, both in early and late life.

Health effects from combustion ultrafine particles: consistent observations from controlled human exposure studies

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The human health effects from exposure to ultrafine particles are challenging to assess. The exposure has complex spatial, temporal, multi-source and co-pollutant variables and the health indicators have multiple etiologies, besides the inter-individual variation in susceptibility. Controlled human exposure studies are a particular study design that allows investigating short-term effects from combustion particles, consisting of experiments where human volunteers agree to be intentionally exposed to pollutants, in a controlled short-term scenario, to provide information on biological changes caused by the exposure. These studies are usually performed in an exposure chamber with controlled air composition, ventilation, temperature, and humidity conditions. Particles are generated by combustion and/or concentration processes or resuspension, and forced into the chamber also allowing the control of mixtures and co-exposures. We revisited the strengths and limitations of this study design and reviewed and compared 64 controlled exposure studies following exposure of a total of 1338 healthy, non-smoking subjects to combustion-generated particles, including 12 chamber studies on wood smoke, 33 on diesel exhaust, 15 on concentrated ambient particles and 4 on indoor carbonaceous sources (candle burning, cooking and printing). Besides different combustion source and generation conditions for the same source, the studies differ by design (crossover or sequential), protocol definitions (with or without moderate physical activity), administrated doses and duration of exposure, as well as the assessed biomarkers and functional markers and methods used in the assessment. The most common markers assessed were inflammation, vascular function and lung function, with less studies focusing on neurotoxicity, arrhythmia and genotoxicity. Consistency of effects (observed in ≥75% of the studies from the same source and involving more than 50 study subjects) were observed after exposure to concentrated ambient particles (vascular function, heart rate variability, arrhythmia and oxidative stress) and diesel exhaust (airway inflammation and vascular function).

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Differential impact of biogenic and anthropogenic secondary organic aerosol compounds adsorbed on soot particles in lung cell models at the air-liquid interface

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BACKGROUND & MOTIVATION: Secondary organic aerosols (SOA) formed from anthropogenic or natural gaseous precursors are substantially contributing to the ambient $PM_{2.5}$ burden, which is known to correlate with adverse human health effects [1]. However, our knowledge is mostly limited to the effects of collected airborne particles under submerged exposure conditions rather than the direct deposition and interaction of aerosols with cell cultures [2]. Moreover, a direct link between single aerosol compounds and their health hazards remains largely unknown [3, 4]. Therefore, we were interested in differentiating the toxicological effects of combustion-derived soot particles (SP) from the effects induced by the soot photochemically aged together with a biogenic (β-pinene) or an anthropogenic (naphthalene) volatile organic compound in two different lung cell models exposed at the air-liquid interface (ALI).

METHODS: Monoculture of lung epithelial cells (A549) and a co-culture model with A549 and endothelial cells (EA.hy926) were exposed at the ALI for 4 h to different aerosol concentrations of pure SP or a photochemically-aged mixture of primary combustion SP and β-pinene ($SOA_{\beta PIN}$-SP) or naphthalene (SOA_{NAP}-SP). The internally mixed soot/SOA particles were comprehensively characterized in terms of their physical and chemical properties. We conducted toxicity tests to determine cytotoxicity, intracellular oxidative stress, primary- and secondary-genotoxicity as well as inflammatory and angiogenic effects.

RESULTS & DISCUSSION: Both investigated SOA types caused significant toxicological effects, while the nano-sized soot cores alone showed only minor toxic effects under the current experimental settings. The toxicological assays furthermore indicated greater adverse effects of SOA_{NAP}-SP compared with $SOA_{\beta PIN}$-SP in both cell models. At the functional level, we showed that SOA_{NAP}-SP augments the secretion of e.g. malondialdehyde and interleukin-8, and may induce the activation of endothelial cells in the co-culture system. This activation was confirmed by comet assay suggesting secondary genotoxicity and an increased angiogenic potential. Chemical characterization of PM revealed distinct qualitative differences in the composition of the two secondary aerosol types. It is shown that SOA-compounds can increase the toxicity of primary SP, which are ubiquitous in inhabited and wildfire influenced areas. Aromatic precursors, such as naphthalene caused the formation more oxidized, more aromatic SOA of higher oxidation potential with higher toxicity compared to an aliphatic precursors, such as β-pinene. The influence of atmospheric chemistry on the chemical PM composition thus can play a crucial role for the adverse health outcome of emissions.

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Exposure-response functions relating intensity and duration of ambient traffic-related air pollution to systolic blood pressure

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Exposure to traffic-related air pollution (TRAP) is associated with adverse health outcomes, including elevated blood pressure and hypertension. However, the nature of the exposure-response functions for these associations is not well established. We recently published a three-exposure, three-period crossover trial where we showed that using portable high-efficiency particulate air (HEPA) filters to reduce indoor infiltration of TRAP was effective at preventing short-term increases in systolic blood pressure (SBP). We measured particle number concentrations (PNC) and black carbon (BC) concentrations continuously during the trial, and we measured participant blood pressures every 10 minutes. In the present study, we derive exposure-response functions that incorporate both intensity and duration of PNC and BC.

A total of 1514 SBP measures arising from 77 participants were analyzed. The average age of the participants was 60 years, 79% were female, 77% were Asian, and 17% were White. None had serious health conditions. The average SBP was 120 mmHg, the average PNC was 13,000 particles/cm^3 (range 860 to 99,000 particles/cm^3), and the average BC concentration was 450 ng/m^3 (range 16 to 2700 ng/m^3). Linear mixed models were used to fit logarithmic relationships between PNC and SBP and between BC and SBP. The models included the variable 'time from study entry' as a measure of exposure duration and a random intercept to account for the interdependence of multiple SBPs recorded from each participant. The resulting models, yielding statistically significant regression coefficients ($p < 0.001$), can be written (and displayed in Figures 1 and 2) as: $SBP = 101.22 + 0.98 \log_e PNC + 2.38 \log_e TIME$ and $SBP = 104.97 + 0.93 \log_e BC + 2.32 \log_e TIME$. The similarity in coefficients between the PNC and BC models can be explained by the relatively high correlation between these two measures of exposure (Pearson correlation = 0.80). Although both models describe a progressive dampening of the increase in SBP for increasing intensities and durations of exposure, duration of exposure has a greater effect on SBP than does intensity; a 1 mmHg increase in SBP is related to an approximate 53% increase in duration but a 185% increase in intensity.

In summary, the results of the present study suggest exposure-response functions for PNC and SBP, and for BC and SBP, that are logarithmically dependent on two dimensions of exposure, intensity and duration.

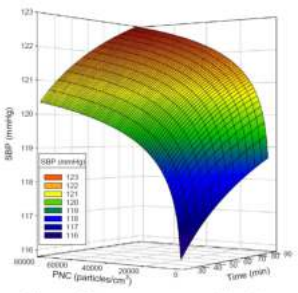

Figure 1. Exposure-response relationship between intensity and duration of PNC and SBP.

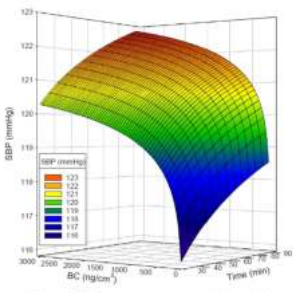

Figure 2. Exposure-response relationship between intensity and duration of BC and SBP.

Development of a new photothermal interferometer for the in-situ measurement of carbonaceous aerosols

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Filter-based instruments are commonly used for the determination of ambient aerosol light absorption. With the assumption of a specific light absorption efficiency, the mass concentration of the light-absorbing carbonaceous particles, particularly black carbon (BC) can be determined. These methods have the advantage of being simple and robust, but the properties of the aerosol change upon deposition in the filter. These methods are prone to measurement artefacts, such as filter loading effects, increased transparency of the filter due to wetting in elevated humidity conditions, and cross-sensitivity to aerosol light scattering.
In-situ methods such as photoacoustics and photothermal interferometry (Moosmüller and Arnott, 1996; Sedlacek, 2006) measure light absorption of aerosols directly in their natural suspended state. They provide an alternative to traditional filter-based instruments. Measurements performed with these techniques are independent of aerosol light scattering and loading effects. Resonant photoacoustic measurements, however, are susceptible to changes in the resonant frequency caused by environmental influences, and suffer from measurement artefacts when absorbing gases are present. In a photothermal interferometer, the aerosols are drawn through a set of measurement chambers where a pump laser illuminates the sample. A small part of the laser light is absorbed by the sample and heat is transferred to the air, locally increasing the temperature, reducing the density and the refractive index. This perturbation is measured with a sensitive interferometer: the change of the refractive index in one arm of the interferometer relative to the unperturbed arm is measured as a change in phase. The pump laser is modulated and the signal at this modulation frequency increases the measured signal.

We present our latest development of a new photothermal interferometry setup (Visser et al., 2020). The use of only one single laser beam allows for a compact optical set-up and significantly easier alignment compared to standard dual-beam photothermal interferometers, making it ideal for field measurements. Due to a unique configuration of the reference interferometer arm, light absorption by aerosols can be determined directly – even in the presence of light-absorbing gases. The instrument can be calibrated directly with light-absorbing gases, such as NO_2, and can be used to calibrate other light absorption instruments. Our current detection limit (1 σ, 120s averaging time) is ~40 ng/m^3 BC.
In new projects, we are investigating the possibility of further significant miniaturization of our existing setup. A unique kind of photothermal interferometer is currently realized using optical fibers and waveguides. Such a miniaturized sensor has many key advantages: It will be smaller, lighter and cheaper than existing BC instruments, which are expensive and rather immobile. The miniaturization into a fully integrated optical circuit board will make it also less susceptible to external vibrations and misalignment. Our goal is to develop a novel miniaturized PTI instrument that can be used airborne to detect the local distribution of BC particles with high temporal resolution.

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Calibration of Black Carbon Instruments with the CPMA-Electrometer Reference Mass Standard (CERMS)

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A wide range of instruments employing a number of different measurement principles report the mass concentration of black carbon (soot). The reported values depend both on the characteristics of the source particles used in the calibration and the reference method applied, neither of which are unique or traceable. The limitations associated with the reference method are avoided by using the centrifugal particle mass analyser (CPMA)–electrometer reference mass standard (CERMS) technique. All aspects of the CERMS technique are traceable to the SI, resulting in a traceable mass standard similar in principle to the calibration for CPCs. The CERMS is capable of classifying and measuring *in-situ* reference mass concentrations down to levels below 1 µg/m^3 and in real-time (~1 Hz). Additional advantages of the CERMS are its low uncertainty (~3 %) and its measurement of a well-defined quantity: total post-CPMA suspended PM mass. The classification of the particles is not affected by particle morphology or composition. When presented with a nonvolatile black carbon (BC) aerosol for calibration, CERMS may be used to calibrate instruments for the measurement of BC mass concentration.

Results for calibration of a number of instruments, including those based on photoacoustic (PAX and MSS), attenuation (CAPS PMssa), and laser-induced incandescence (LII 300) principles are presented. A number of sources for BC are evaluated, including laboratory soot generators (MISG and miniCAST) and engine emissions (gas turbine and diesel). The results show that the BC instrument responses for different sources do vary, as a result of differences in the particle morphology, internal structure, or composition. As thermal-optical analysis (TOA) to determine elemental carbon (EC) mass is the reference method for the mass concentration measurement of aircraft gas turbine engine nonvolatile particulate matter (nvPM) emissions, a comparison between EC mass from TOA and nvPM mass from CERMS is also discussed and closure for mass is demonstrated under carefully-controlled conditions [1]. The sensitivity of the CERMS is shown by characterizing the limits of detection of the LII 300 and PAX instruments. The results support the use of CERMS with well-characterized BC sources to provide reference mass concentrations for the calibration of instruments measuring BC mass concentrations.

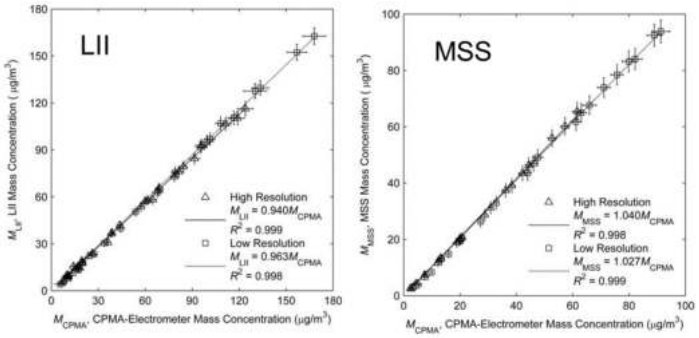

Figure 1. Demonstration of using the CERMS technique to determine calibration factors for the LII 300 (left) and MSS (right) instruments with a black carbon aerosol.

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Performance of the new continuous carbonaceous aerosol measurement system FATCAT during long term unattended measurement campaigns

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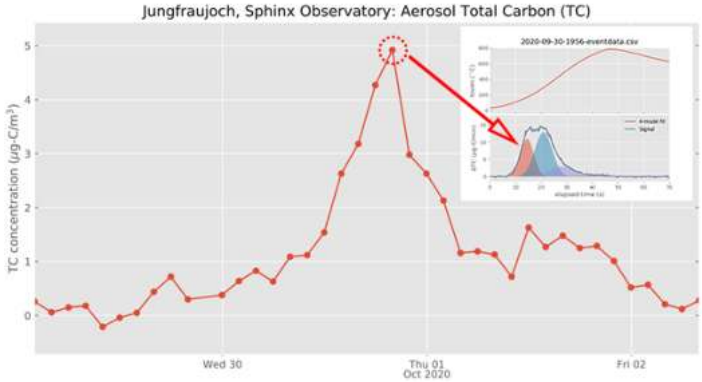
Total aerosol carbonaceous mass (TC) is a major constituent of atmospheric fine aerosol particle mass. However, this fraction is generally not continuously monitored with an adequate time resolution. Adding a TC measurement is crucial to complement the existing measurement programs for a comprehensive interpretation impact of aerosols on our climate. To fill this gap, we developed the “fast thermal carbon totalizator” (FATCAT), a carbonaceous aerosol measurement system for long-term monitoring of TC. FATCAT has been deployed since 2019 at different measurement sites, including Zurich (urban roadside), Windisch (urban background), Payerne (suburban background), and the Sphinx observatory at the Jungfraujoch global GAW station (JFJ; above the planetary boundary layer) for unsupervised long-term measurement campaigns. FATCAT collects a sample on a sinter metallic filter, and subsequently heats it to 800°C under an oxidizing atmosphere. The fast-heating cycle of 50 seconds allows a low limit of detection (LoD) of 0.2 µg of carbon (µg-C).

We will discuss our experience during the first two year of continuous TC measurements and the possibility of using our instrument to distinguish carbonaceous aerosol from different source using its fast thermograms. This unique feature allows us to identify source specific fingerprints. For instance, several high TC episodes during September 2020 at the JFJ station show the typical pattern for biomass combustion. With the identified fingerprint and back trajectories, these episodes were attributed to long-range transported emissions from Californian wildfires (see figure). In general, refractory, crystalline carbon from, e.g., fossil fuel combustion evolves at high temperatures, whereas aerosol particles from, e.g., biomass burning sources containing more amorphous organic carbon decompose and evolve at lower temperatures. The dataset generated by our instrument and post-analysis data products represent an improvement to the available measurement inventory. It can also serve as quality control for other measurement systems. Prominently, measurements of eBC via MAAP or Aethalometer and organic mass using ToF-ACSM require calibration and are susceptible to systematic errors. TC measurement data can be used in parallel for these devices as a quality check but also to warrant total carbon mass closure and reduce systematic biases.



This work has been supported by global atmosphere watch (GAW) and by MeteoSwiss through the GAW-CH Science Projects 2018-2021 The authors acknowledge the support from MeteoSwiss, the city of Zurich, EMPA, University of Bern, and the Paul Scherrer Institute (PSI Switzerland) for their assistance during the measurement campaigns.

Effects of ambient CO_2 and H_2O on soot formation in n-dodecane spray combustion

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In this study, large eddy simulation (LES) is performed to investigate the effects of ambient carbon dioxide (CO_2) and water (H_2O) on the soot formation in an n-dodecane spray flame. A two-equation soot model, in which acetylene (C_2H_2) is set as the soot precursor and surface growth species, while OH is selected as the one of the soot oxidizers, is implemented here. The ambient oxygen (O_2) level and temperature are fixed at 15% (mole basis) and 900K, respectively. The predicted ignition delay, lift-off length, and soot distributions show good agreement with experimental data. The effects of ambient CO_2 and H_2O on the soot formation can be separated into thermal and chemical effects. For the thermal effects, the ambient CO_2 and H_2O enhance the formation of C_2H_2 but reduce the formation of OH radicals by lowering the flame temperature. This leads to a higher soot mass formed. Conversely, the ambient CO_2 and H_2O reduce the soot formation due to their chemical effects. The reaction $CH_2^* + CO_2 \leftrightarrow CH_2O + CO$ is found to be main pathway for reducing C_2H_2 formation when the ambient CO_2 is present. The ambient H_2O results in a lower C_2H_2 mass formed due to a higher amount of OH radicals produced. As a result, these collectively lead to a lower soot mass formed.

Towards the reduction of brake and tire emissions: The Zero Emission Drive Unit (ZEDU-1)

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Technological advances, standardized testing, and legislation regarding engine-related emissions have been effective in reducing particulate emissions from newly registered vehicles. In order to achieve further progress here, it is important to conduct similar efforts at vehicle components that receive less attention in the context of particulate matter reduction but are relevant for any form of propulsion. This includes tires, which play a major role in the generation of coarse particles, and brakes, which emit high levels of ultrafine particles during operation. The goal of the Zero Emission Drive Unit (ZEDU-1) research project is to design and characterize a first-generation drive axle that enables zero-emission driving with high efficiency and effectiveness while remaining suitable for everyday use. Specifically, this includes a new concept for the effective minimized release of particles in the environment via absorption of tire abrasion and the development of a brake system that is free of fine dust emission even at full braking performance. From a range of braking technology, a viscous brake was selected for the battery-electric ZEDU demonstrator vehicle, to prevent any particulate emissions to the environment. Other technologies to reduce brake emissions such as new coatings of disc brakes, and the use of an induction hybrid brake are also investigated in the scope of the project.

For the technology evaluation, particle emissions will be investigated not only based on the total mass but the entire particle size distribution. This includes online measurements of particles from the range of ultrafine up to PM10 with an EEPS, OPS, and a DMS500. For offline chemical analysis, particles are size-selectively collected with an ELPI+. Besides, the fraction of non-volatile particles is determined by using a catalytic stripper.

A multi-stage test concept is planned to ensure reliable and realistic investigation of the non-exhaust particle emissions: First, a baseline for brake and tire abrasion is determined using a conventional electric vehicle (BMW i3). For this purpose, a specific enclosure was designed that allows mobile measurements on the vehicle and direct source identification. Chassis dynamometer measurements with the WLTP cycle and the new WLTP braking cycle allow reproducible measurements, as well as an estimation of various influencing factors. Second, the developed and tested sampling setup is used to evaluate the new brake coatings. Particulate emissions are measured under identical conditions and directly compared with emissions from a conventional brake. Finally, the emissions of the ZEDU demonstrator vehicle will be determined on a chassis dynamometer under the same driving cycles and environmental conditions. Additional mobile measurements are planned during runs on a test site.

This work is carried out as part of the ZEDU-1 project and supported by the Baden-Württemberg Ministry of Economics, Labour and Housing.

Handheld Emission Particle Counter for testing diesel particle filters of off-road engines

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Introduction & Background

In Switzerland, construction machines and other off road diesel engines need to have a diesel particulate filter to minimize the exposure of on-site personnel to carcinogenic diesel soot particles. To test the correct function of these filters at construction sites and other locations, a mobile diesel soot sensor is needed, which is able to measure the particle number concentration of soot particles directly at the exhaust. This sensor has to fulfill the specifications of the Ordinance of the Federal Department of Justice and Police on Exhaust Gas Measuring Devices for Internal Combustion Engines (VAMV SR 941.242). The Aerosol Group of the Institute for Sensors and Electronics at the University of Applied Science and Arts, Northwestern Switzerland (FHNW) developed such a device, the Handheld Emission Particle Counter (HEPaC). The HEPaC is based on the Partector2 by naneos LLC. It has been certified by METAS.

Methodology and Results

The HEPaC is a diffusion charging sensor, consisting of a heavily modified Partector2. The result of the measurement is transferred to a tablet PC to generate protected PDF reports. To avoid problems with condensation or nucleation, the exhaust aerosol enters the sensor via a probe followed by an evaporation tube, which is heated to 195°C. The sensor itself is heated to 55°C. This enables the fulfilment of the tetracontane test, i.e. evaporating at least 95% of 30nm tetracontane particles up to a particle number concentration of 10^5 pt/cm³. The heated evaporation tube is followed by the diffusion charger, which charges the soot particles constantly, followed by a pulsed electrostatic precipitator, switched between two voltage levels. The resulting periodically changing charge induces a pulsed current (fA) on arrival in a faraday cage, which is measured by an electrometer. To control the aerosol flow rate a differential pressure sensor and a nozzle are used in this sensor. When all operational parameters like diffusion current, pulsed precipitator voltage and flowrate are correct, the amplitude of the electrometer current is directly proportional to the particle number concentration of the aerosol. The battery operation time is approximately 3 hours.

Conclusions

The HEPaC is a METAS certified lightweight sensor, which allows mobile and simple measurements of particle number concentrations of construction machines directly at the construction site. The sensor implementation follows the protocol for Swiss regulation SR 941.242. It works up to a number concentration of 5’000’000 particles/cm³ with a CPC like counting efficiency curve. The efficiency versus particle size also fulfills the requirements of the Dutch PTI regulations and the suggestion by PTB for PTI.

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Effects of overlapping in the evaluation of volume and surface area of complex soot aggregates in flames

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The release of ultrafine soot particles to the atmosphere can cause detrimental effects on the environment for example by influencing the formation of clouds and/or by changing the radiative properties of the atmosphere [1]. In most numerical works, soot particles are typically considered either spherical in some numerical simulations or fractal aggregates in advanced codes that neglect the overlapping between primary particles. These approximations lead to significant deviations in the surface area and volume of soot particles, which in turn can cause large uncertainties in modeling soot physicochemical processes depending on particle sizes, such as collision rates and surface reaction rates (both surface growth and oxidation).

Fractal-like aggregates representative of flame-made soot (fractal dimension 1.78, prefactor 1.30, and 2-1000 number of spherical monodisperse primary particles) are generated by using a recently developed sequential algorithm called FracVAL [2]. Subsequently, primary particles are enlarged uniformly to induce a desired level of overlap in the 0-90% range. The volume and surface area of these aggregates are then calculated using the accurate but expensive (in CPU time) SBL library [3]. Based on these calculations, different equations to approximate their volume and surface area are proposed. In addition, a new method to estimate the population average overlapping coefficient is introduced. This requires the total number of collisions to be known at all times. As a test case, the method is implemented to predict the total surface area and volume of soot aggregates in an ethylene (C/O=0.94) laminar premixed flame [4].

The proposed equations extend the works of [4,5] to determine total soot volume and surface area in time. Current expressions are accurate for aggregates of any size relevant to soot particles produced in flames at atmospheric pressure. Under the test case conditions (premixed flame), neglecting primary particle overlapping leads to a maximum overestimation of soot particle total volume and surface area of 91 and 218%, respectively. The proposed approach exhibits a maximum error of 0.6% for total volume and 5.75% for total surface area. This method only requires the total number of collisions in time, the surface growth rate (the rate of change in primary particle diameter), and the number of primary particles per aggregate. All of these parameters are readily available in most population balance simulations making its implementation in existing codes straightforward. The proposed method can also be used in future Monte Carlo or Langevin Dynamics discrete element simulations to potentially reduce the CPU time associated with the evaluation of aggregate volume and surface area.

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In vitro toxicity of airborne emissions from combustion of graphene nanoplatelet-enabled epoxy nanocomposites

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Combustion is one of the processes occurring at the material's end of life. The combustion process can release the embedded nanoparticles from the nanocomposite's matrix and might transform the nanoparticle's properties[1]. An increasing use of graphene nanoplatelets (GNPs) as an additive in commercial products raises concerns about the potential risks of the released particles, especially human exposure to airborne fraction of the released GNPs since inhalation is one of the major exposure routes. Despite many studies about the release of nanoparticles induced by combustion [2], [3], the hazard assessment of the GNPs released from the combustion is still limited. Therefore, this study aims to characterize the particulate and gaseous emissions from the combustion of neat epoxy (EP) and GNP-enable epoxy composite (EP-GNP) and evaluate the biological effects of the emissions on human alveolar epithelial cells (A549) cultivated at air-liquid interface for up to 96 h after the combustion exposure. The particle modal sizes of the emissions from both EP and EP-GNP were in the respirable range of 4 µm. We found volatile organic compounds and polycyclic aromatic hydrocarbons (PAHs) in the soots and gases from the emissions. After the treatment with the emissions from both EP and EP-GNP, the quantification of lactate dehydrogenase suggested no adverse effects on cell membrane integrity. Cells treated with the emissions from EP-GNP, but not EP, showed a significant reduction in mitochondrial activity at 24 h time point. However, this effect was transient and values recovered to those of filtered air controls after 96 h. Release of the inflammatory chemokines/cytokines MCP-1 and GM-CSF was increased at 24 h time point after exposure to both EP and EP-GNP, while the values were dropped to control levels at 96 h. Expression of CYP1A1 gene, associated with metabolic activation of PAHs, strongly increased with exposure to both EP and EP-GNP at both time points. Exposure to EP-GNP emissions caused slightly higher CYP1A1 expression at 24 h than exposure to EP emissions. Our results reveal potential enhanced toxicity from GNP nanofillers, which should be considered in future risk assessment studies and the safe design and use of GNP-enabled nanocomposites.

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Combustion-generated carbonaceous urban atmospheric UFPs: Fuel optimisation in transport and decentralized electricity economies to minimise their production and impact

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34% carbonaceous ultrafine particles (UFPs) in the atmosphere come from road transport [1]. Their toxicity generally increases as the particle size decreases. They are found in urban atmospheres worldwide [2] and aggregate to form larger assemblies. Dobbins concluded from TEM that flames and diesel engines generated nascent single nanoparticles (NPs) (2<d<10nm; 1.8< C/H <2.2) that were precursors to carbonaceous fractal chain aggregates (5< C/H <10) and nanotubes [3], both with PAHs adsorbed thereon [4]. Lawther saw a relationship between air pollution and the incidence of bronchitis [5] and heart risks rise 38% for women who live within 49m of busy roads [6]. Nano-organic carbon (NOC) particles and hydrophobic soot NPs are emitted more from diesel- than gasoline-fuelled vehicles (i.e. the ratio of soot:NOC rises from 0.5 (gasoline engines) to 3.0 (diesel engines)). Here we describe the minimisation of carbonaceous UFP *and* CO_2 emission from generator and vehicle engines (using H_2-O_2injectants and modified fuels (i.e. bio-esters in gasoline engines or water-in-biodiesel emulsions in diesel engines (see Figure 1))) and describe control of their primary particle size/fractality/PAH passenger load/hydrophobicity/toxicity/impact. Carbon is of the seven elements that have changed the world [7]; this work is focussed on making its nanoparticle impact (as we transition to a zero-carbon future) only positive.

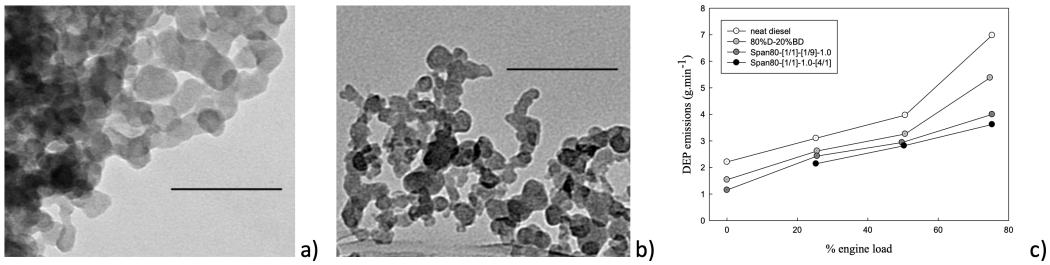

Fig.1 TEM micrographs of diesel emitted carbonaceous UFP particles with various tested fuels at 75% load: (a) diesel and (b) water-in-biodiesel emulsion (scale bars = 100nm). (c) Formation of DEPs is lower in Span 80 stabilised water-in-diesel/biodiesel emulsions at all loads.

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