

Ultrafine particle concentrations in Kloten near the Zurich Airport (Switzerland)

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Ultrafine particles (UFP) comprise the smallest particle fraction of atmospheric particulate matter with particle diameters of less than 100 nm. In recent years, studies in the surroundings of international airports have shown that air traffic can be an important source of UFP. We report on UFP measurements performed since February 2019 in a residential area in Kloten (urban background), a site located 1 km east of Zurich Airport and about 500 m east of highway A51 (approximate traffic activity of 100'000 vehicles per day). Both the airport and the highway are located to the west of the measurement site, making it challenging to distinguish the contributions from both potential major sources of UFP at this location. UFP at the site in Kloten are measured as number size distribution of particles with diameters between 10 and 100 nm. Reported UFP number concentrations ($PN_{[10-100nm]}$) cover this size range.

Between February 2019 and February 2020, the mean concentration of UFP was about 20'000 particles per cm^3 , a value that corresponds to typical UFP concentrations in Switzerland at roadside locations directly adjacent to busy roads. However, in Kloten the UFP concentration depends strongly on wind direction, wind speed and time of day. For example, the average UFP concentration at wind directions from west to northwest was for the February 2019 to February 2020 period about 40'000 particles per cm^3 and therefore three times higher than for winds prevailing from southeast (about 14'000 particles per cm^3). Beside total UFP concentration, the mean particle diameter in Kloten also shows a strong dependence on wind direction with the smallest particle diameters for winds from western and northwestern directions, i.e. from the directions in which both the airport and the highway A51 are located. The strongly enhanced UFP concentrations at winds from west to northwest, as well as the fact that the measured average UFP concentration is clearly higher than at typical Swiss roadside sites indicates significant contributions from air traffic at the site in Kloten. However, the quantitative determination of the air traffic contribution to total UFP concentration requires more elaborated analysis of the measured particle size distributions and consideration of information from the scientific literature. For the measurement site in Kloten, we determined the contribution of air traffic at Zurich Airport (landing and takeoff) to the total UFP concentration in 2019 to be 40%, based on the distribution of concentration peaks combined with wind conditions. While air traffic is a dominating source of UFP at this site, it is of minor importance for the concentration of the regulated air pollutants NO_2 , $PM_{2.5}$ and PM_{10} .

The intervention measures to reduce transmission of the SARS-CoV-2 virus implemented in March 2020 resulted in a marked decline of road traffic on highway A51 and had a dramatic effect on the air traffic activity at Zurich Airport. Consequently, the UFP concentrations at the site in Kloten strongly declined. The change in UFP concentrations was much stronger linked to the change in air traffic activities compared to road traffic, confirming the influence of air traffic on the level of UFP in the studied residential area in Kloten.

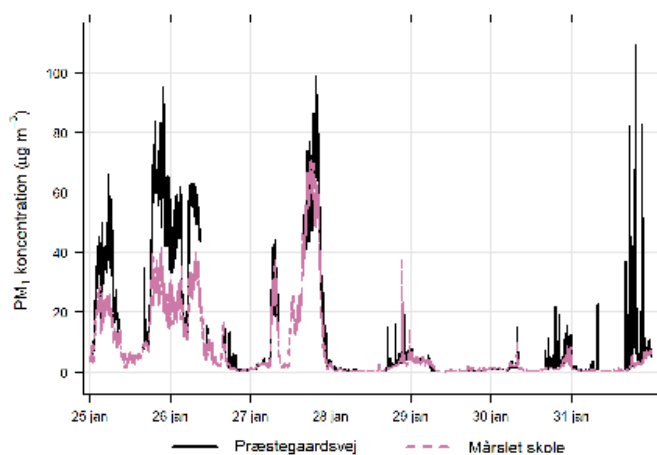
Clean air test-zone - reduction of emissions from wood stoves

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Today, wood stoves and boilers are estimated to account for 65-70% of particle and black carbon emissions in Denmark. At the same time, the legislation relates exclusively to type approval of stoves in the laboratory, which rarely reflects real-life emissions when used in private homes. A new Danish lighthouse project (2020-2023) demonstrates modern stoves and emission-reducing technology under real-life conditions in a dedicated residential neighborhood (test zone) in the municipality of Aarhus, Denmark. The objective is, from a cost-benefit perspective, to identify measures that provide most impact on real-life emission reduction from wood stoves, including automatic control stoves, low-emission stoves, new types of chimneys, draft boosters, ESP filters and catalysts. Additionally, digital apps can help the consumer firing more correctly. In order to evaluate effect of the various approaches, distributed low-cost sensors continuously quantify local air pollution levels (PM, CO, NO_x, temperature, wind direction and speed). Through developed data algorithms, we expect to identify which point sources are actually problematic and which are acceptable. The distributed sensors are supported by dedicated point source emission measurements (PM, PN, NO_x, CO, CO₂, OGC).

The dedicated test-zone, with around 25 actively participating households, was established in the city of Mårslet in autumn 2020. Several outdoor and indoor sensors (Leapcraft) are mounted and continuously logging PM and gases in the 20/21 heating season. Present measurements in the 20/21 heating season will serve as a reference background for technical interventions targeting wood stove emission reductions during the coming two heating seasons. One of the main challenges in the data analysis is separating the wood stove signal from other sources. Present efforts involves pattern recognition and comparing with parameters such as background level, wind direction, relative humidity and chimney temperature. As an example, below is shown the PM₁₀ concentration measured in a lamppost in the test zone (Præstegårdsvej) and background (Mårslet school) for a week by the end of January. In general, the concentration measured at Præstegårdsvej is significantly higher than at Mårslet school, very likely due to local sources of particles, of which wood-burning stoves are expected to make up a significant part. This is supported by the fact that the high concentrations occur in the evening and at night, where an increased use of wood-burning stoves can be expected. At ETH 2021 a more detailed analyses will be given on the above, expected to include a preliminary quantification of the signal from wood stoves - as well as results from reference measurements on individual chimneys in the test zone.



Macro tracer model as a technique for source apportionment of particulate matter in Krakow agglomeration - an optimization approach

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Introduction:

Receptor models are mathematical procedures used to identify the sources of pollutant emissions and to estimate their contribution in the overall balance of atmospheric aerosol emissions based on measurements of the concentrations of aerosol components, without the need to carry out an inventory of emission sources or data describing meteorological conditions. The concept of a *macro tracer* model consists in determining a chemical compound - an indicator - specific to a given source. The basis of the model is the work of P. Lenschow et al. who took the first steps to match a given indicator compound with a given particulate matter emitter¹. The *macro tracer* model was then developed in Austria by scientists from Vienna University of Technology^{2,3}. The assumptions of the model had to be optimized in order to implement this model to the region where coal is the main source of energy. New factors have been calculated on the basis of source profiles obtained in preliminary studies presented by Katarzyna Szramowiat-Sala et al.⁴. Afterwards, the optimized *macro tracer* model was used for the identification of emission sources of atmospheric aerosols from Krakow agglomeration.

Results & Conclusions:

The *macro tracer* model enabled to obtain reliable results on the contributions of individual sources of atmospheric aerosols in the Krakow agglomeration in winter. The optimized model allowed to reconstruct the PM mass with the higher R² linear coefficient than using the F coefficients estimated by Gonçalves et al and Kistler et al. However, to obtain more appropriate results, it is necessary to broaden the research studies on chemical composition of particulate matter from individual sources and to identify the more specific tracer for them.

Acknowledgement:

The work has been partially financed by the subvention project of the AGH UST in Krakow (no. 16.16.210.476) and using infrastructure of the Centre of Energy, AGH UST in Krakow.

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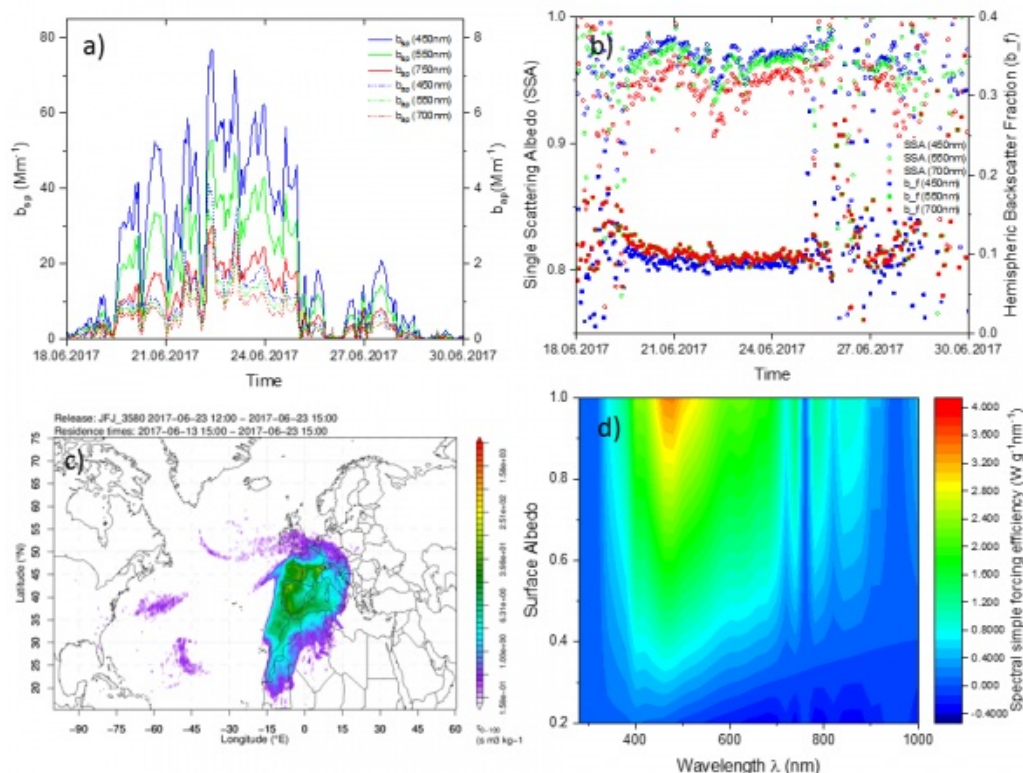
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Occurrence and Radiative Properties of Long-range Transported Wildfire Aerosol Measured at the Jungfrauoch

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Our history has been a story of how we and fire have co-evolved. The same holds for our future. Wildfires are often not “wild” and can be linked to human activity. Hence, some claim that we are living in the “Pyrocene” [1]. Large-scale biomass burning events emit substantial amounts of primary particulate matter (PM) and gaseous secondary PM precursors. Fractions of these emissions can be injected into the free troposphere where they can be transported over long distances and exhibit a stronger radiative forcing efficiency than at ground level [2]. Large light absorbing carbon concentrations in the free troposphere can also alter the vertical temperature profile, leading to a stratification of the atmosphere below with consequences for clouds and precipitation [3]. This work characterizes the optical and microphysical properties of wildfire plumes transported to the Jungfrauoch (JFJ) Station (3571m a.s.l.). Forty-eight wildfire plumes were identified from hourly averaged data for the period between January 2015 and December 2020 using a data filter based on gaseous data and aerosol optical properties. The Figure below shows the scattering and absorption coefficients (a), the SSA and backscatter fraction (b), the source sensitivities (c) and the Mie theory derived simple forcing efficiency (d) of the most significant plume event in the period analysed, that originated from the Pedrógão Grande wildfire in Portugal in June 2017. Ongoing work validates the optical data filter with particle size distribution-, gaseous- and source sensitivities- data. In addition, plume occurrence frequency and plume microphysical properties will be presented at the conference.



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The UNREAL (Unveiling nucleation mechanism in aircraft engine exhaust and its link with fuel composition) project: Results from simulation chamber and reactor experiments

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Aviation emissions are not limited to greenhouse gases like CO₂ but include other gases as well, such as nitrogen oxides (NO_x) or sulfur oxides (SO_x) and volatile and non-volatile particulate matter (vPM and nvPM respectively). Sulfuric acid formed in the engine exhaust seems to be linked to the formation of vPM. However, the amount of sulfur present in the fuel converted to sulfuric acid in the exhaust is too small to explain the amount of vPM observed and organic species seem to play a key role in their formation [1]. The UNREAL project aims at studying at the molecular level the different mechanisms of new particle formation from the exhausts of aircraft engines fed by different fuels (Figure 1). We used a Combustion Aerosol STandard (CAST) generator especially designed to work with liquid fuel to generate the emissions from different fuels (from the standard Jet A-1 to 100% Sustainable Aviation Fuel (SAF). Total, or only gas-phase, emissions were injected in both, an atmospheric simulation chamber (CESAM) and a Potential Aerosol Mass Oxidation Flow Reactor (PAM-OFR) for aging. Both systems used in parallel provide a point of comparison between in near-real time and hours-long time chamber aging approaches. The chemical and physical evolution of primary and secondary CAST emissions, were monitored by different instruments to measure particle number, mass concentrations as well their size distribution, and the chemical composition of both, the particulate and gas phases. In addition to online techniques, samples were collected to study off line, the gas and aerosol chemical composition at molecular level by mass spectrometry.

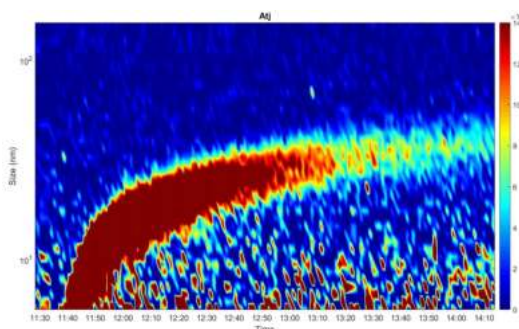


Figure 1. Nucleation event from emissions of 100 % Atj fuel in reaction with OH radical when only gases were introduced into the atmospheric chamber.

This work benefited from the support of the project UNREAL ANR-18-CE22-0019 of the French National Research Agency (ANR).

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Impact of fuel composition on primary and secondary aeronautic emissions: gaseous and particulate chemical characterization at molecular level

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One of the actual concerns of aviation industry is to reduce its impact on climate and air quality using for example Sustainable Aviation Fuels (SAF, [1]). As part of the UNREAL Project [2], the objective of this work was to study and compare chemically, at a molecular level, gaseous and particulate phases of primary and secondary emissions from various aircraft fuels, from the standard Jet A-1 to 100 % SAF (Alcohol to Jet). Related emissions from a liquid CAST burner [3] were injected into an atmospheric chamber (CESAM), and in parallel, into a Potential Aerosol Mass Oxidation Flow Reactor (PAM-OFR) to age the emissions (OH reactivity in both cases). To collect these fresh and aged particles, a system with two quartz fiber filters was used: the Front Filter (FF) traps the particulate phase while the Back Filter (BF) is coated with activated carbon to retain the gas phase [4]. Samples were analyzed with a Two-Step Laser Mass Spectrometry (L2MS) technique to study the chemical composition of emissions, in particular Polycyclic Aromatic Compounds (PAC) and sulfur such as SO₂, SO₃ or H₂SO₄. The chemical composition of the filter-deposited samples is compared to on-line measurements performed by an Aerosol Mass Spectrometer and an Aerosol Chemical Speciation Monitor for particles, and a Proton Transfer Reaction Mass Spectrometer (PTRMS) for gaseous species. A comparison of the primary and aged emission molecular chemical fingerprints obtained, as well as between both atmospheric reactors (CESAM vs PAM-OFR), will be proposed.

This work benefited from the support of the project UNREAL ANR-18-CE22-0019 of the French National Research Agency (ANR).

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Particle Emissions from Aircraft Gas Turbines: A Coarse Size Mode from Low Emission Engines

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This presentation is about coarse particle emissions from a commercial aircraft gas turbine engine. These particles were found in a series of test campaigns focused on understanding and improving methodology for measuring non-volatile particles from these engines. Measuring or even finding coarse particles was unexpected. Before describing these measurements, some background is useful.

Motor vehicle engine exhaust particle size distributions typically show three distinct modes: a nucleation mode between about 3 and 30 nm diameter consisting mainly of semi-volatile material, an accumulation mode between about 30 and 300 nm diameter consisting mainly of carbonaceous aggregates, and a coarse mode consisting of larger mechanically generated particles - from oil atomization and re-entrainment from in-cylinder and exhaust surfaces.

Measurement of particles from commercial aircraft gas turbine engines is much more challenging than from ground-based vehicles with exhaust temperatures as high as 900 C or more and exhaust velocities approaching Mach 1. This necessitates the use of very long sampling lines that complicate particle measurements. These lines tend to adsorb much of the semi-volatile material and suppress the formation of a nucleation mode. The carbonaceous aggregates in the accumulation mode are much smaller than from typical piston engines with geometric mean diameters in the 15 to 50 nm range. Coarse mode particles are also smaller and here we define them as particles larger than 150 nm.

The results reported here were observed during a series of measurement campaigns conducted by the U. S. Environmental Protection Agency in collaboration with the U. S. Air Force's Arnold Engineering Development Complex. The main purpose of these campaigns was to refine methodology for measurement of non-volatile particles from aircraft engines. A General Electric J-85 turbojet running with a range of test conditions and fuels was used as the particle source. Particle size and concentration measurements were made using a range of instruments. The focus of these studies was measurement of accumulation mode soot aggregates. Compared to piston engines there is less opportunity for the exhaust to interact with surfaces and there are no piston rings to atomize oil so coarse particles were not expected to be an issue.

Coarse particle size measurements were made using a range of instruments. Four measurement campaigns were conducted. In the first, a pair of TSI SMPSs operating in the sizing range of 8 to 300 nm were used. In the second, SMPSs in the 6 to 225 nm range were used. In the third, SMPSs were still operated in the 6 to 225 nm range but a TSI EEPS that operated in the 6 to 500 nm range was added. In the fourth and final study, three SMPSs were operated in the 6 to 225 nm range, and a fourth in the 15 to 690 nm range. The TSI EEPS was again used in the 6 to 500 nm range and a Cambustion DMS500 operating in the 5 to 1000 nm range was added.

The significance of the coarse mode was not noticed until the fourth campaign when volume fractions, $(V \text{ above } 150 \text{ nm})/V_{\text{total}}$, V_{150}/V of nearly 50% were observed in some tests. After that, all test campaigns were reexamined. In most case, V_{150}/V varied inversely with engine load and total volume (mass) emissions. Biofuels, hydrotreated camelina oil blends with Jet-A generally produced higher V_{150}/V .

Test results will be presented, and possible sources and formation mechanisms will be discussed.

Assessment of global particle number emissions from shipping and effect of scrubbers

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Globally, ship exhausts are the main source of particles over large areas in open seas and coastlines, and the particles emitted both contribute to impaired air-quality and have climatic effects. Combustion aerosols indirectly affect climate by influencing the formation of clouds and their light scattering properties. In order to accurately estimate the climate-forcing impacts of ship emissions, particle number (PN) and size distribution (PSD) of the emitted aerosols are important, further to total particle mass (PM). From 2020 onwards, ships are forbidden to burn fuel with sulphur content over 0.5%, unless they apply scrubbers for SO₂ abatement.

Particle measurements were conducted in laboratory on 1.6MW marine engine and 6 different fuels with varying properties and sulfur contents were tested at low and high load points. In addition, measurements were conducted on-board a cruise ship, before and after scrubber, from the exhaust lines of two main engines (ME1 and ME2, applying SCR). Identical sampling setup was applied in all campaigns, consisting of a porous tube diluter together with residence time chamber and ejector diluter, which simulates atmospheric dilution conditions [1]. PN and PSDs were studied by scanning mobility particle sizer and condensation particle counters (CPC). To study the effect of scrubber on non-volatile particles, a catalytic stripper was applied. The obtained PN emission factors were compared to ship plume observations conducted on the Finnish coastline, by chasing ship plumes by aircraft. STEAM ship emission model was applied to assess the globally distributed PN emissions from international shipping and estimate the influence of the sulfur regulation.

The PN emission factors for different fuels varied between 1.38-5.83×10¹⁶ 1/kgfuel [2]. The scrubber efficiently removed volatile particles in nucleation mode size range and reduction was larger with engine applying SCR [3]. The global PN emissions from shipping are localized close to coastal lines and busy port areas, but significant emissions exist also on open seas and oceans. The global annual PN produced by marine shipping was 1.2×10²⁸ (±0.34×10²⁸) particles in 2016, which is of same magnitude with total anthropogenic PN emissions in continental areas [4]. The potential to reduce global PN emissions from shipping depends strongly on the adopted technology mix, and is possible with wide adoption of natural gas and scrubbers, but no significant decrease is expected if heavy fuel oil is mainly replaced by low sulfur residual fuels.

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Analysis of the influence of marine fuels on particle emissions from ships

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In contrast to the extensive requirements for land-based particulate and fine dust emissions, the maritime sector is still in its infancy and has not yet been able to provide any global limit value regulations. It can be assumed that this topic will become more and more important in the future. With the last tightening of the IMO regulations on January 1st, 2020 (sulfur content in fuel $\leq 0.5\%$ or use of an exhaust gas cleaning system outside of the ECAs), a change in the composition of the fleet emissions is to be expected. In this context, the joint project SAARUS was launched at the University of Rostock, with the aim to investigate ship-based emissions and to reduce them through optimized and expanded exhaust gas cleaning. In addition to reducing SO_x emissions, the focus is on separating fine particles that measure smaller than 2.5 μm (PM_{2.5}). In particular, the health-endangering fine dust fractions (aerosols) with particle diameters below 1 μm are only slightly reduced by conventional wet scrubbers. The approach to further decrease the particle load is therefore to use the scrubber as an optimized particle prefilter in order to create the boundary conditions for downstream filter technologies to be tested.

In this context, an extensive measurement campaign with six different fuels available on the market took place on a medium-speed single-cylinder research engine, which is representative of the maritime sector and located at the Chair for Piston Engines and Internal Combustion Engines. As part of the investigations, the fuel-based changes in emissions and the combustion behavior of a hydrogenated vegetable oil (HVO), a MGO, a limit-compliant HFO (sulfur content $\leq 0.5\%$), a standard HFO (sulfur content 2.4%) and two highly aromatic heavy fuel oils (sulfur content 0.06% and 1.3%) are analyzed. The following measurement methods were used to characterize the particle emissions: gravimetric filter analyzes, tapered element oscillating microbalance (TEOM), scanning mobility particle sizer (SMPS), Pegasor particle sensor, online single particle mass spectrometry (SPMS), filter sampling and two-dimensional gas chromatography / mass spectrometry (GCxGC-TOFMS), high-resolution mass spectrometry (HRMS for organic matter) and inductively coupled plasma / mass spectrometry (ICP-MS for elements).

The focus of the article is on the presentation of the most important findings of this measurement campaign. In addition to a comparison of the properties of the fuels examined, their effects on the particle load in terms of concentration, size distribution and chemical composition are discussed. In addition, the simulation approach for particle separation in the scrubber and the approaches for separating fine particles measuring smaller than 2.5 μm (PM_{2.5}) as well as the harmful fine dust fractions (aerosols) with particle diameters below 1 μm are presented in an outlook.

Evaluating the performance of a particle counting sensor based on continuous-wave laser-induced incandescence

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Introduction & Background:

We investigate the influencing factors for the performance of an optical particle sensor based on the principle of continuous-wave laser-induced incandescence (cw LII). The sensor's main target is the determination of the number concentrations for soot particles with a detection limit regarding the particles' diameter of below 100 nm. In our previous work, we presented a functional compact sensor demonstrator using the focused light of a laser diode for the heating of soot nanoparticles and a silicon photomultiplier for the detection of LII events [1]. With a comparable laboratory setup, the use of such a sensor to reliably determine the soot particle number concentration was shown [2]. For the introduction of the size-dependent count rates, a SMPS was used to measure the mean diameters of the investigated size distributions. In future, this information can be directly deduced from the registered LII signal shapes.

Methodology:

The choice of suitable optical components such as laser source, lenses and mirrors was supported by a systematic study of the associated laser light distribution in the focus and the filter efficiencies regarding the LII transmission and stray laser light suppression. Based on these results, functional demonstrators were built. One critical value to assess the sensor's performance is the determination of the minimum detectable particle size. By using an indirect method that compares the created particle size distribution to the events registered with the LII sensor, an indication about this figure-of-merit can be derived. We compare the influence of different parameters like the laser power densities, particle velocities, and signal detection methods on the detection limit.

Results & Conclusions:

Through experiments with a variation of the particle distribution generated by a miniCAST soot generator, the indirect method to determine the detection limit helps to understand the cause-effect relationships influencing the sensor performance. The obtained results support further improvements of the sensor setup. The use of near-monodisperse particle distributions allows to improve the accuracy of the determination of the sensor's detection limit. For the most promising setup, this approach led to a detection limit well below 100 nm. Measurements with various particle sources showed a broad range of derived size distributions. This includes the use of a miniCAST, a kerosene soot point lamp, and a diesel engine. The evaluation of the registered signal peaks reveals a dependency between particle size and signal shape, which opens the possibility to directly correct the monitored count rates.

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Dual particle counter for measuring simultaneously automotive exhaust solid particle number emissions larger than 10 nm and 23 nm

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>Introduction and Background

The Solid Particle Number (SPN) emissions of Light Duty (LD) vehicles are regulated in Europe since 2014. The tests are conducted by sampling diluted exhaust from a Constant Volume Sampling (CVS) system. Current legislation (UNECE R83) requires a Volatile Particle Remover (VPR) which consists of an Evaporation Tube (ET) and a Condensation Particle Counter (CPC) with a cut-off of 23 nm. The latest Amendment of the Global Technical Regulation for certifying LD vehicles specifies a catalytic stripper (CS) instead of the ET and a CPC with a cut-off of 10 nm instead of 23 nm.

Methodology

A dual line SPN system (APC xApp) was developed to allow for parallel measurements with an AVL 10 nm and an AVL 23 nm CPC. The APC xApp was employed in parallel to a system fully compliant with the UNECE R83 regulation (APC^{plus} [1]) in measurements of Gasoline Direct Injection (G-DI) LD vehicles from a CVS. The APC^{plus} employs an ET and a 23 nm AVL CPC. The vehicles were tested under the Worldwide harmonized Light vehicles Test Procedure (WLTP) and other driving protocols. The study investigates the influence of the CS on the > 23 nm, as well as the ratio of the SPN 23 to SPN 10 emissions.

Results and Conclusions

The ratio SPN 10 to SPN 23 ranged from 1.1 to 1.5, in line with already reported literature figures [2, 3]. The SPN >23 nm differences between APC^{plus} (ET) and APC xApp (CS) were found to be confined within $\pm 10\%$. The observed differences were within the measurement uncertainty (e.g. originating from calibration [1]), and showed no dependence on the ratio of 10 to 23 nm concentrations (and therefore size of emitted particles). The results verify that the use of a CS has no effect on SPN > 23 nm, in good agreement with what anticipated from the efficiency curves of the CPCs and APCs.

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Super Polluter IDentifiER (SPIDER) - a tool for on-road detection of vehicles that contribute disproportionately to the vehicle fleet emissions

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Traffic is a diverse and disperse source of air pollution. The amount of pollutants emitted depends on vehicle parameters; the engine type and displacement, the exhaust after-treatment system, maintenance status, as well as traffic conditions, topography, driver behavior and the weather conditions. Different statistical analyses and measurement approaches have been employed to evaluate traffic emissions; these vary in complexity in terms of describing traffic activity and emission factor (EF) determination. The EF determination methods performed on-road in real driving conditions have been described as less precise than the dynamometer studies because the tests are not as repeatable due to the absence of standard cycles and additional uncontrolled parameters, such as environmental or traffic conditions, driver behavior or highly transient operations [1]. Their advantage over dynamometer/laboratory measurements is that, over a short period of time, emissions of many different in-use vehicles can be measured and EF distributions for different vehicle categories can be obtained. There have been several studies measuring EF of individual vehicles using different approaches [1-2] and references therein.

We have repeated the on-road measurement campaign using the chasing method as described in Ježek et al. [2] and determined EF of BC and nitrogen oxides (NO_x) from 435 individual vehicles of different types and particle number (PN) EF of 145 different type vehicles. We have acquired technical data from the national registry information about the measured vehicles and analyzed the data according to vehicle type, age and fuel used. The data-processing method was re-evaluated and optimized, especially from the point of view of background concentration determination, which can represent an important source of uncertainty.

The vehicles were firstly grouped to three categories - diesel cars, gasoline cars and goods vehicles, and then according to age and introduction of new European emission standards. Significant reductions in BC EF with introduction of EURO5 and EURO6 emission standards in diesel cars and goods vehicles groups were observed. The reduction in NO_x EF for diesel cars was small and found only in the EURO6 group. We observed the reduction in NO_x EF in goods vehicles and gasoline cars categories. PN EF were also significantly reduced in all three vehicle categories. We will compare measurement results to the results of Ježek et al. [2] and the contribution of super emitters in the main three vehicle groups. We will demonstrate that the optimized SPIDER method can be used in real-world to determine supper emitters, and the associated reduction of fleet emissions, if these supper emitters were eliminated from the fleet.

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Soot particle ice nucleation ability dependence on their volatile contentK. Gao^{1,3}, C. Zhou^{1*}, H. Koch², Z. Kanji^{3*}

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Aviation traffic in the upper troposphere can significantly influence the climate via CO₂ and non-CO₂ emissions. Soot particles, as important non-CO₂ particulate emission, interacting with water vapor in the atmosphere can act as ice nucleating particles (INPs), and are able to impact the aviation contrail evolution and further cirrus clouds formation. The latter has a larger coverage than contrails globally and plays an important role in modulating the Earth's radiative forcing and climate. Therefore, the ice nucleation ability of aviation soot particles is of particular interest.

In this study, the dependence of soot particle ice nucleation ability on volatile content (organics) is investigated at mixed-phase and cirrus cloud conditions. Four types of soot sample can be classified into two classes, first, two types of nonporous propane flame soot used as aviation soot surrogates (Ess and Vasilatou, 2018; Marhaba et al., 2019) and second, two types of porous commercial carbon black with different levels of volatile composition. The results show that thermal denuding at 573 K in a pure N₂ and a compressed air (N₂ + O₂) atmosphere modifies the soot particle ice nucleation abilities in a non-systematic manner between 218 and 233 K. However, the same treatments have pronounced effects on the nonporous propane flame soot particle ice nucleation activities. In particular, our results show that organic carbon removal by thermal denuding in compressed air suppresses the ice nucleation ability of organic lean propane flame soot particles at T < 233 K. On the other hand, organic rich propane flame soot particles treated at the same conditions have a tendency for an enhanced ice nucleation activity. This is contrary to the previous study that reports organic carbon can considerably suppress propane soot particle ice nucleation (Mohler et al., 2005).

In brief, this laboratory study demonstrates that soot particle ice nucleation activities show dependence on particle treatment which potentially affects their properties, inclusion of chemical composition and surface oxidation condition changes, as well as aggregate structure compaction. Thus, transport and ageing in the atmosphere that perturb the chemical properties of soot particle should be considered to accurately reflect ice nucleation activities. The findings about propane flame soot are of significance to evaluate the impacts of aviation emissions and contrail evolution on the climate. In particular, the role of volatile substances like organic carbon coating and its effects on soot particle ice nucleation ability deserve further investigation.

Acknowledgement This work is supported by the Chinese Scholarship Council (Grant No. 201906020041) and the the Experimental Atmospheric Physics Professorship, ETH Zurich.

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Detection and quantification of combustion-derived particles in aqueous media: towards the development of a diagnostic biomedical assay

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Abstract:

The detection and quantification of combustion-derived particles (CDPs) such as carbonaceous particulate matter (PM) in biological samples is important to understand their toxicity. Different techniques, such as absorption photometry and laser-induced incandescence, used for measuring CDP concentrations in gaseous environments, cannot be employed for the label-free detection of CDPs in biological samples. Hence, there is a need for the development of diagnostic tools for selective and label-free detection of CDPs to evaluate the exposure at the level of individual persons. Recently, we have shown that CDPs could be detected in biological samples based on non-incandescence-related white light (WL) emission under illumination with femtosecond (fs) pulsed near-infrared (NIR) lasers using a multiphoton microscope.[1-4]

In this work, we study the effects of stirring and concentrations of suspensions on the label-free detection and quantification of particles under illumination with fs pulsed NIR laser. We performed measurements on CDP suspensions with different concentrations in ultrapure water and buffered medium as exemplary conditions for wet biomedical samples. We observe that the number of detected particles increases linearly with the concentration of the suspension. This opens the ways for label-free quantification of CDPs in liquid biopsies.

In addition to CDPs, ambient PM also includes other types of nanoparticles such as silica and metal oxides. This increases the likelihood of interference from other common nanoparticles in the detection of CDPs based on WL emission. In our recent study[5], we could observe that the WL emitted by the CDPs under illumination with fs pulsed NIR lasers is unique and was not observed for other common nanoparticles. Hence, interference from other types of common nanoparticles based on WL emission is not expected when detecting CDPs in aqueous media.

We believe that these results are a step towards the development of diagnostic biomedical assays for direct and label-free detection of CDPs at the level of individual persons.

Keywords: particulate matter, combustion-derived particles, nanoparticles, femtosecond pulsed laser, white light emission

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Effects of Outdoor Smoke Events on Indoor air Quality

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It is known that climate change has greatly enhanced the probability of extreme wildfires. While people spend most of the time at home or at the workplace, little is investigated how indoor air filtering systems are performing under intensive outdoor smoke conditions. For this aim, particle number size distribution and concentration in a size range 0.5 – 18 μm and equivalent black carbon (eBC) mass concentration were measured in outdoor and indoor office air simultaneously.

7-wavelength Aethalometer (Magee Scientific AE31) and Aerodynamic Particle Sizer (APS) (TSI 3321) were deployed in the Center for Physical Sciences and Technology (FTMC) campus located in the urban background environment in Lithuania from 30th September to 6th October 2020. Since all parameters were evaluated for indoor and outdoor air during the event (fire) and non-event (no fire) days, the air filtration system's performance during a long-range wildfire event were evaluated. Outdoor and indoor office air was investigated through various metrics: particle size distribution, $\text{PM}_{2.5}$, PM_{10} , and eBC mass concentrations. Filters selectivity for different eBC sources (biomass burning versus traffic) and chemical composition of carbonaceous aerosol particles (eBC versus brown carbon (BrC)) was tested as well. It was found that the coarse particle number concentration was found to be 14 times higher in comparison with clean periods in indoor air. The smoke event resulted in twice higher indoor and outdoor eBC mass concentrations. Because of lower removal rate for small particles, eBC had higher contribution to total $\text{PM}_{2.5}$ mass concentration in indoor air than in outdoor air.

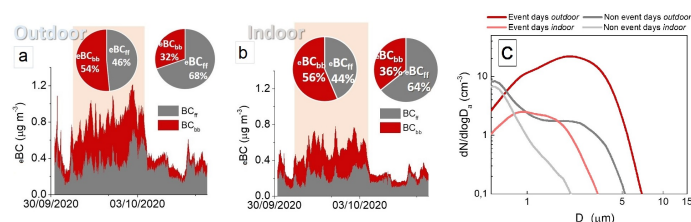


Fig 1. Time series of eBC_{ff} and eBC_{bb} mass concentrations in outdoor (a) and indoor (b) air. The pink areas mark the smoke event. Pie charts represent contributions of each parameter during non-event and event days. Particle number size distribution in outdoor and indoor air for the event and non-event days (c).

Acknowledgments. This research was funded by a grant (No. S-MIP-20-28) from the Research Council of Lithuania.

Organic analysis of aircraft engine smoke number filter samples with thermal-optical carbon analysis and thermal desorption-gas chromatography/mass spectrometry

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From atmospheric measurement near an airport runway [1], we have recently found that jet engine lubrication oil [2] is a key component of aircraft exhaust nanoparticles [3]. However, it is uncertain where oil nanoparticles are emitted or formed in aircraft jet engine exhaust. In this study, we aimed to examine the organic composition of jet engine exhaust bulk particulate matter (PM) collected with a smoke meter, especially to see whether those PM samples have lubrication oil signature. The smoke number (SN) paper filter samples, collected at SR Technics, Zürich Airport, Switzerland in 2019–2020, were used. From carbon analysis, the volatile organic carbon (OC) concentrations ($\mu\text{gC cm}^{-2}\text{-filter}$) in the gray-colored SN filter samples were very low but significantly higher than the blank filters. The roughly estimated volatile OC concentrations in the undiluted exhaust was $1,200 \mu\text{gC m}^{-3}$ for the gray-colored samples. In the chromatograms of thermal-desorption gas chromatography/mass spectrometry (TD-GC/MS), oil marker compounds (tricresyl phosphate and likely fatty acid esters of pentaerythritol) were clearly and consistently detected from the gray-colored samples, and were not detected from the blank filters (Fig. 1). The mass spectra of the oil marker compounds in the gray-colored samples matched well with those of the jet engine lubrication oil. From the gray-colored samples, strong signals of $\text{C}_{24}\text{-C}_{30}$ *n*-alkanes and some polycyclic aromatic hydrocarbons (PAHs) were also detected.

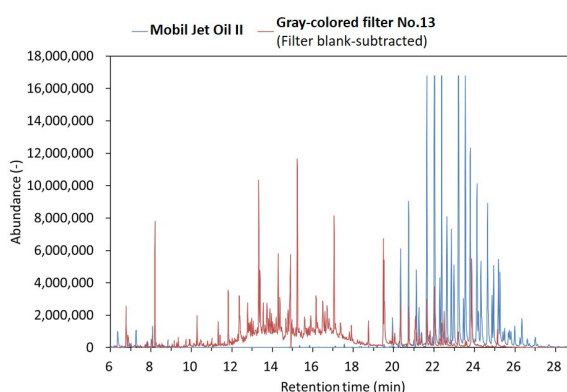


Fig. 1 Overlapped TD-GC/MS mass chromatograms (m/z 85: major ions of hydrocarbons) of the gray-colored SN filter sample and jet engine lubrication oil.

This work was supported by the Environment Research and Technology Development Fund (JPMEERF20205004) of the Ministry of the Environment, Japan, the NIES Research Funding (Type B), and the Scientific Exchanges grant of Swiss National Science Foundation (IZSEZO_198063). The work of the Swiss team was funded by the Swiss Federal Office of Civil Aviation (FOCA) projects AGEAIR SFLV 2017-030 and AGEAIR 2 SFLV 2018-048.

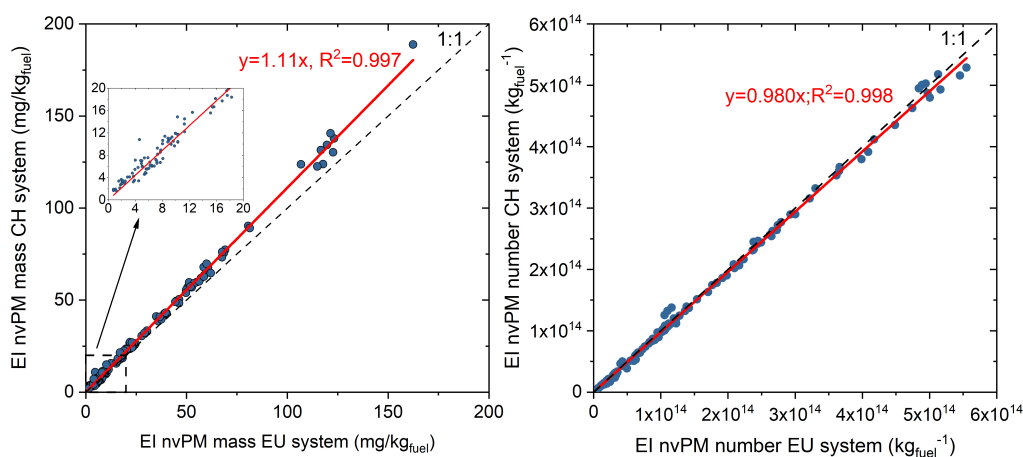
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Intercomparison of two reference sampling and measurement systems for aircraft engine non-volatile PM using a small-scale RQL combustor rig burning conventional and sustainable aviation fuels

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Aircraft gas turbine engines directly emit non-volatile PM (nvPM) with electrical mobility diameters mostly below 100 nm, affecting local air quality and the climate. To mitigate nvPM pollution, the International Civil Aviation Organization (ICAO) has introduced certification standards for nvPM mass and number emissions of aircraft turbine engines with rated thrust above 26.7 kN. However, further work is needed to characterise and reduce nvPM emissions measurement uncertainty and particle loss correction to provide better estimations of engine exit concentrations for airport emission inventories. As a part of the first campaign of the Horizon 2020 funded project RAPTOR, two nvPM reference sampling and measurement systems (Swiss and EU) were operated in parallel and sampled exhaust from a small-scale aero-engine rich-burn quick-quench lean-burn (RQL) combustor rig burning a range of conventional and sustainable aviation fuels at multiple rig operating conditions. Additional particle size measurements were performed using a TSI SMPS with a catalytic stripper in the Swiss system and a Cambustion DMS-500 fast spectrometer in the EU system.



The preliminary results of this study show good agreement between the two systems for the nvPM number and mass emission indices (EIs). At low nvPM mass concentrations, larger discrepancies were observed between the two systems because of the shedding of particles accumulated in the PM1 cyclones installed in each system. At specific rig conditions, the RQL combustor produced bi-modal particle size distributions with no volatile fraction which were similarly captured by both the SMPS and the DMS-500. It was found that the standardised particle loss correction methodology (only requiring measured nvPM mass and number) was inaccurate when particle size distributions were bi-modal and/or low nvPM mass concentration when compared with particle loss correction estimated using measured particle size distributions. This study will lead to a better understanding of the uncertainties of the regulatory nvPM data and supply data to improve the measurement methodology and more accurate prediction of aircraft engine nvPM emissions released into the environment.

A Silver Particle Generator for PN-PEMS Calibration and Validation

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There currently exist few commercial options for the consistent and reliable generation of solid aerosol particles in the 1-50 nm size range. These options include spark generators, tube furnaces, electrosprays, and gas burners utilising diffusion flames. Thus, there is a need for additional options for a simple-to-use, particle generator capable of producing sufficiently high concentrations of small, solid aerosol particles. Uses for such a device include the calibration of condensation particle counters, measuring filtration efficiencies, and the calibration and complex measurements made possible with specialist aerosol equipment, including cutting-edge emissions measurements such as PN-PEMS. The efficiency of PN-PEMS is checked with monodisperse aerosol, and linearity is checked usually with polydisperse aerosol in order to reach high concentration levels [1]. Silver particles can be used as a proxy for soot particles and given their single elemental composition this approach may offer reduced uncertainties to other generation techniques. Tube furnaces can be used to generate silver nanoparticles for calibrated CPCs, but aside from the size, cost, and inconvenience of a tube furnace, it is challenging to generate repeatable concentrations and size distributions from a typical tube furnace.

Here we present the characterisation of a new Silver Particle Generator, capable of producing sufficiently high concentrations of particles in the 1 - 50 nm size range. Data presented include CPC calibration, and repeatability measurements across its operational range. This novel Silver Particle Generator solves several key aspects associated with the production of metallic nanoparticle aerosols. By fixing the location and surface of metal exposed to the supplied gas stream, the stability of the nanoparticles produced is greatly enhanced compared to a typical tube furnace. Further, day-to-day variability is greatly reduced, in both concentration and size domains. The SPG has been finely tuned to generate a repeatable solid aerosol size distribution for each set point, and thermal cycling has been optimised, such that the device is operational from standby in under 10 minutes. Importantly, our novel and patented design allows for a more compact device that allows for lower energy consumption.

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