Available online at www.sciencedirect.com

ScienceDirect

Transportation Research Procedia 72 (2023) 248–255

Transportation Research Procedia

www.elsevier.com/locate/procedia

Transport Research Arena (TRA) Conference

Nanoparticle emissions from the transport sector: health and policy impacts – the nPETS concept

Ulf Olofsson[a], Ellen Bergseth[a*], Jens Wahlström[b], Karine Elihn[c], Hanna Karlsson[d], Haibo Chen[e], Dimitris Margaritis[f], Zisis Samaras[g], Anastasios Kontses[g], Fulvio Amato[h], Benjamin Piña[h], Jose Portugal[h], Barend van Drooge[h], Sharon Ridolfo[h], Xavier Querol[h], Mara Leonardi[i], Christer Johansson[c,j], Magnuz Engardt[j], Isabel Hermandez[k], Emilio Benfenati[l], Andrea Colombo[l], Jorma Keskinen[m], Ana Juarez[c], Yezhe Lyu[b], Minghui Tu[a]

[a]*KTH Royal Institute of Technology, Department of Machine Design, Brinellvägen 83, 100 44 Stockholm, Sweden*
[b]*Lund University, Ole Römers väg 1, Lund 221 00, Sweden*
[c]*Stockholm University, Department of Environmental Science, Svante Arrhenius väg 8, Stockholm 106 91, Sweden*
[d]*Karolinska Institute, Environmental Medicine, Nobels väg 13, Stockholm 17177, Sweden*
[e]*University of Leeds, Institute for Transport Studies, 34-40 University Road, Leeds LS2 9JT, UK*
[f]*CERTH, 6thkm Charilaou-Thermi rd, Thermi GR 57001, Thessaloniki, Greece*
[g]*Aristotle University of Thessaloniki, Laboratory of Applied Thermodynamics, 3is Septemvriou, Thessaloniki 541 24, Greece*
[h]*Institute of Environmental Assessment and Water Research (IDÆA), Spanish National Research Council (CSIC), Barcelona 08034, Spain*
[i]*BREMBO Spa, Viale Europa, Stezzano, Bergamo 24040, Italy*
[j]*SLB-analys, Environment and Health Administration, Box 8136, 104 20 Stockholm, Sweden*
[k]*Catalonia, Carrer de Provença 204-208, Barcelona 080 36, Spain*
[l]*Mario Negri, Via Mario Negri 2, Milano 20156, Italy*
[m]*Tampere University, Aerosol Physics Laboratory, P.O. Box 692, FI-33014 Tampere University, Finland*

Abstract

Road, rail, air, and sea transport generate a major fraction of outdoor ultrafine particles. However, there is no common methodology for comparable sub 100 nm particle emissions measurement. This paper presents the nPETS (grant agreement No 954377) concept to understand and mitigate the effects of emerging non-regulated nanoparticle emissions. This paper presents the concept and selected results. For example, nucleation and condensation mechanisms occur more frequently in the urban background site, leading to new particle formation, while mostly fresh emissions are measured in the traffic site.

Keywords: Nanoparticles; Toxicology effect; Social acceptance; Air quality; Health and quality of life; Impacts of health measures in mobility

* Corresponding author. Tel.: +46 8 7907383
E-mail address: bergseth@kth.se

10.1016/j.trpro.2023.11.401

1. Introduction

This paper presents the nPETS research concept to understand and mitigate the effects of emerging non-regulated nanoparticle emissions on public health, combining aerosol science, toxicology, transport policy studies, and engineering science. All disciplines are highly interdependent. The consortium consists of seven technology-facing universities, three world-leading European research institutes in aerosol monitoring and modelling, one European company with state-of-the-art techniques to measure vehicle brake emissions, and two local administrations (Stockholm and Barcelona). The three-year project started in June 2021.

Although the formation of molecular clusters takes place almost everywhere and all the time, new particle formation (NPF) events (i.e., rapid bursts of particles) have a spatial and temporal correlation with ultra-fine particle (UFP) emissions, according to Rönkkö et al. (2017). NPF events are most observed during rush hours in urban areas, as Flemming et al. (2019) reported. Traffic is also a source of precursors for secondary particle formation in cities, as presented by Kerminen et al. (2018). These precursors are emitted in the gas phase but are transformed into the particulate phase in the atmosphere through oxidation processes. Fig. 1 illustrates NPF transformation from source to the urban environment, including the time aspect. The first particle transformations occur from the time the particles are released from the source until they are initially oxidised in the atmosphere. Primary particles are the ones coming directly from the source (e.g., measured in engine lab), and secondary particles are primary particles that have been exposed to UV light (e.g., outdoor at street level/near the source). The fresh particle formations occur between primary and secondary but are not exposed to an outdoor climate like in a road or rail tunnel. Aged aerosol with secondary aerosol contributions will be studied in the urban background.

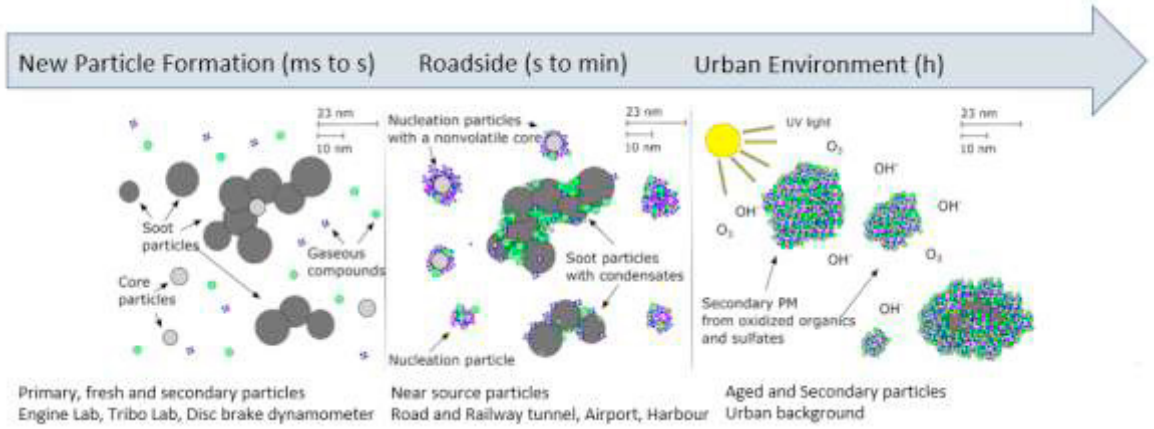

Fig. 1. Illustration of New Particle Formation (NPF) transformation from source to urban environment and in which set-up we expect to measure the various particle formations. The arrow illustrates the time aspect. Adapted from Simonen et al. (2017) and the project DownToTen's final review meeting (2020).

nPETS proposes a cross-sectoral concept of the activities to assess the emissions of nanoparticles from all transport modes, see Fig 2. Quantification and monitoring of sub 100 nm emissions are made in controlled laboratory environments and field campaigns using the same instruments to ensure comparable measurement results. Particles collected from these studies will be characterised by size, morphology, and chemical composition. Toxicology studies will be performed on sampled particles from filters. Also, in situ cell exposure will be performed using an Air Liquid Interface (ALI) system presented by Latvala et al. (2016). The project also includes a review of the current knowledge on the impact of sub 100 nm emissions on toxicology and epidemiology. The gained knowledge will serve as input for a review of future sub 100 nm emission health policies and their economic impact.

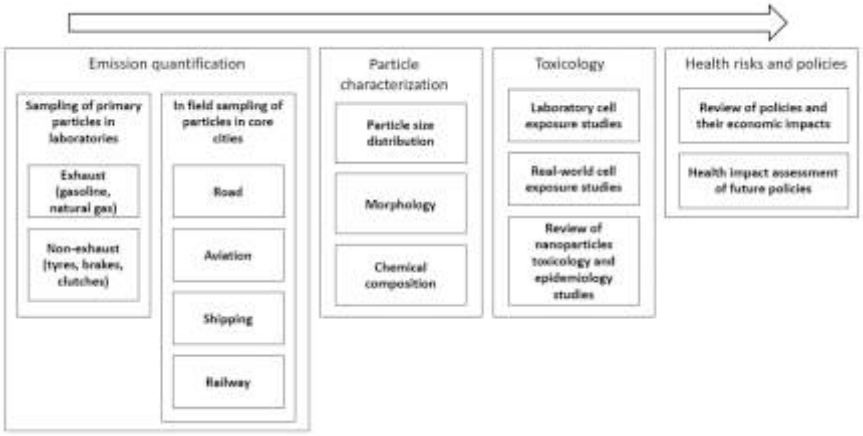

Fig. 2. The nPETS concept, in nPETS Grant Agreement (2021). The arrow suggests the timeline of the project activities.

The following chapters present an overview and selected results, mainly from emission quantification, that serves as a base for further activities. Chapter 2 introduces the instruments, and chapter 3 introduces the field and lab tests. Chapter 4 describes some initial particle analysis work, and the last chapter mentions European research collaborations.

2. Particle measurements instruments

Since there is no common methodology for comparable measurement of sub 100 nm particle emissions from the transport sector, all quantification and monitoring will be performed with the same instruments to ensure comparable measurement results. The main instruments are hereby described.

The Dekati Gravimetric Impactor (DGI) is a high sample flow rate cascade impactor. The DGI collects size classified particles in four stages with Ø47 mm substrates, analysed gravimetrically or chemically after the measurement. The tiniest particle size fraction is collected on a Ø70 mm filter. Once the mass flow is 100 l/min, the measurement range is between 0.13 and 2.1 µm in aerodynamic diameter. Therefore, the size range of the particles collected on the end filters is approx. <100 nm. During the measurements, greased aluminium filters are placed on the upper four stages, and PTFE filters with 3 µm pores are used as end filters.

The Electrical Low-Pressure Impactor Plus (ELPI+) is used in the nPETS project. It measures particles between 6 nm and 10 µm in aerodynamic diameter using a low-pressure impactor and electrical detection with sensitive electrometers. The detected particles are classified according to aerodynamic diameter into 14 impactor stages. The time resolution is 1 s. This ELPI+ has also been upgraded to an advanced version, which is called High-Resolution ELPI+ (HR-ELPI+). It uses a data inversion algorithm that gives real-time particle number size distribution in up to 500 size classes. During the measurements, greased aluminium filters are placed on the nine stages, and non-greased polycarbonate filters are placed on the finer stages (<100 nm size range).

A mobile Air Liquid Interface (ALI) system has been built and validated in the lab and field. Fig. 3a-c shows the system. Using the ALI system, we mimic the interactions of airborne particles with lung cells. As a result, we provide more knowledge of health risks linked to nanoparticle exposure. In addition, the ALI system enables deposition of un-changed airborne particles, i.e., size, shape, and chemical composition of particles same as in the air – and no need for particle collection. The deposition of particles directly on cultured human lung cells (electrostatic deposition of charged particles) resembles human inhalation of airborne particles. The system has separate chambers and two chambers with filters (cells exposed to a flow of particle-free air). Aerosol is tempered and humidified to keep the cells alive. A Scanning Measurement Particle Sampler (SMPS) before and after the ALI system enables online determination of the cell particle dose and allows testing of cytotoxicity and inflammatory markers.

Later, the project decided to add the general SMPS to get particle size distribution. In addition, Particle Number (PN) concentration will be measured in most cases with a Condensation Particle Counter (CPC). Also, during the tests at the different laboratories and field locations, additional instruments will be used parallel to the above described to

provide more information on the particle and gaseous emissions. The indicative instrument list comprises the Fast Mobility Particle Sizer (FMPS) and the Engine Exhaust Particle Sizer (EEPS) for particle size distribution, additional CPCs sampling from different sampling and conditioning systems to provide information on the PN concentration of solid and volatile species, aethalometers and black carbon instruments, gas analysers for the determination of e.g., NOx, CO and SO2 emissions.

3. Field campaigns and laboratory tests

Each field campaign is estimated for four weeks. Most of the campaigns (Table 1) will cover two seasons. Exhaust particle emissions are tested in a chassis dynamometer and engine lab. Non-exhaust sources from mechanical brakes are studied using a brake dynamometer and disc-brake tribometer. Other non-exhaust sources are tested using tribometers for imitating the clutch, tyre-to-road, wheel-rail, and third-rail contact.

Table 1. Field campaigns, city and type of transport and emission source and test location.

	Road	Rail	Air	Sea	Urban background
Barcelona	Roma Avenue	Valencia metro	Josep Terradellas	Barcelona Harbour	Palau Reial
Stockholm	Söderledstunneln	Stockholm metro			Torkel Knutssonsgatan
Thessaloniki	Egnatia & Ionos Dragoumi		Thessaloniki Airport	Thessaloniki Harbour	Thessaloniki
Milan	Viale Marche	Saronno railway station	Milan Linate Airport		Milano Pascal

3.1. Field campaigns

In Stockholm, the equipment is in a room next to the tunnel for road measurements. Tubes on the wall connect with the tunnel environment, and the measurement devices can continuously sample the air inside the tunnel. The representative devices used on this site are DGI, ALI system (initially validated at the metro station in Stockholm, see Fig. 3a-c), ELPI+, SMPS, NOx, and aethalometer. The measurements have been carried out from November 2021 until March 2022. Average observed concentrations were NOx: 850 µg/m^3, PNC: 85000 /cm^3, BC (880 nm) 4 µg/m^3. However, the airborne particle level on the metro station underground platform is continuously measured using a permanent set-up (Fig. 3d). Instruments are Fidas 200S, DGI, FMPS, NOx and aethalometer. Average concentrations from October 2021 to March 2022 were: PNC: 5000 /cm^3, BC (880 nm) 5 µg/m^3. The urban background measurements are always ongoing, while Differential Mobility Particle Sizer (DMPS) started in February 2022. Average concentrations for the period December 2021 – February 2021, were PNC: 5000 /cm^3, BC (880 nm) 0,4 µg/m^3.

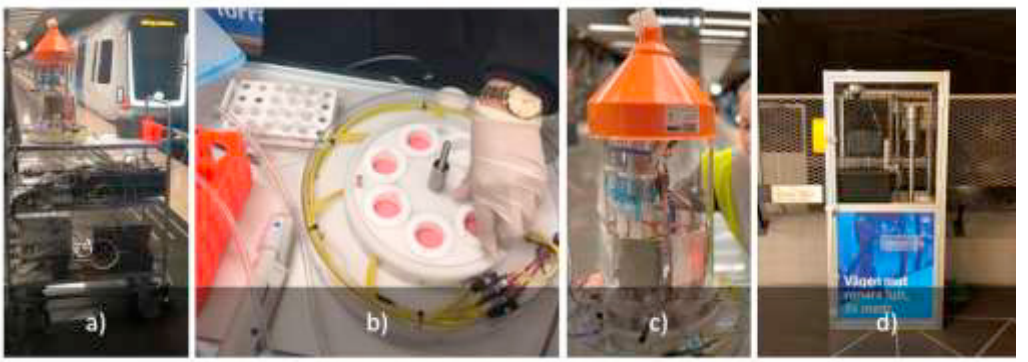

Fig. 3. (a) Photos from when setting up the mobile ALI-system; (b) a close up of the separate chambers and (c) the assembled system. (d) The permanent measurement station at Tekniska Högskolan metro station, Stockholm.

In Barcelona, we have obtained Particle Number Size Distribution (PNSD) both from ELPI+ (at urban background and traffic site) and SMPS (at the urban background site). We have also tested the DGI impactor at both sites, allowing preliminary particle Inductively Coupled Plasma-Mass Spectrometry (ICP-MS), Scanning Electron Microscope (SEM), Transmission Electron Microscopy (TEM) and toxicological analysis on the samples collected with ELPI+ and DGI.

Fig. 4 shows daily averaged mass and number size distributions in a working day using ELPI+ in the urban background (Palau Reial) and the traffic site (Eixample). The urban background site mass had a bimodal distribution, with peaks in the fine particles (D50=0.63 μm) and coarse particles (D50=10 μm). A unimodal mass distribution was observed in the traffic site, with the peak found in the coarsest stage. Most particles were found below 100 nm at both measurement sites regarding number size distribution, although a second smaller peak was at 130 nm. Higher concentrations were observed at the traffic site. The observed differences are attributed to the fact that nucleation and condensation mechanisms occur more frequently in the urban background site leading to new particle formation, while in the traffic site, mostly fresh emissions are measured. Results using the SMPS (not shown here) at the urban background site (Palau Reial) in July-August 2021 and January-February 2022 showed that the number size distribution in the warm and cold seasons differed. Although both distributions were unimodal, differences were observed in the peak size diameter. In summer, the peak is set at 10 nm, while the peak is found at 20 nm in winter. This difference is due to the seasonal variation of the emission sources. In summer, the meteorological conditions favour nucleation, resulting in new particle formation in the range <20 nm, while in winter, fresh traffic emissions that usually peak around 20 nm dominate.

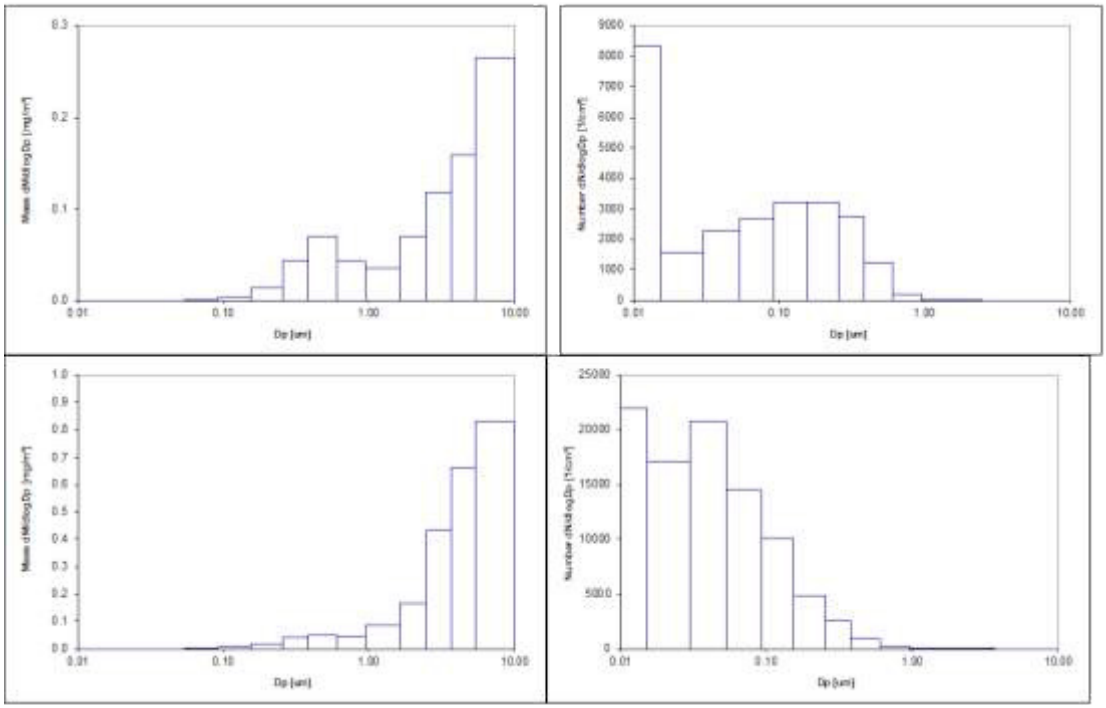

Fig. 4. Example of daily averaged mass size distribution (left) and number size distribution (right) in the urban background site (top) and traffic site (bottom).

3.2. Laboratory testing

Fig. 5 shows the installation on the chassis dynamometer (Fig. 5a) of the latest technology (Euro 6d-temp) gasoline direct injection (GDI) vehicle equipped with a gasoline particulate filter (GPF), and the collected particulate matter

on the DGI substrates (Fig. 5b). Indicative results on solid PN and soot emissions are presented in Fig. 6, while the chemical composition of the collected nanoparticles will be determined in the next step, and the toxicity of the nanoparticles will be evaluated with relevant in vitro testing.

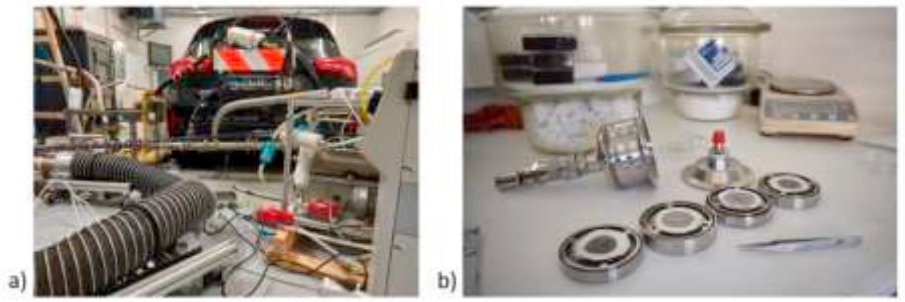

Fig. 5. (a) Installation of vehicle (gasoline direct injection vehicle equipped with a gasoline particulate filter) and measurement equipment at AUTh-Laboratory of Applied Thermodynamics. (b) Substrates of the DGI with collected particles.

Fig. 6 presents the size distribution, PN and soot emissions over two cycles in cold and hot-start conditions. The conditions are: the Worldwide harmonised Light vehicles Test Cycle (WLTC), the current type-approval cycle in Europe; and a combined cycle, a combination of a US06 cycle followed by a BAB130 and another repetition of the US06. The target of such a combined cycle is to include high-load events and dynamic motorway driving. Fig. 6 show that solid PN emissions are well below the current limit (applicable only on WLTC) of 6×10^{11} p/km, while soot emissions remain at low levels in all tests. The cold-start combined cycle has the highest PN emissions. However, a high variation was observed in the hot-start repetition of the same cycle, and the reason is still under investigation. Regarding the particle size distribution, a bimodal distribution is observed in all cases, with a first peak at around 10 nm and the second at about 100 nm, revealing the importance of nanoparticle emissions investigation.

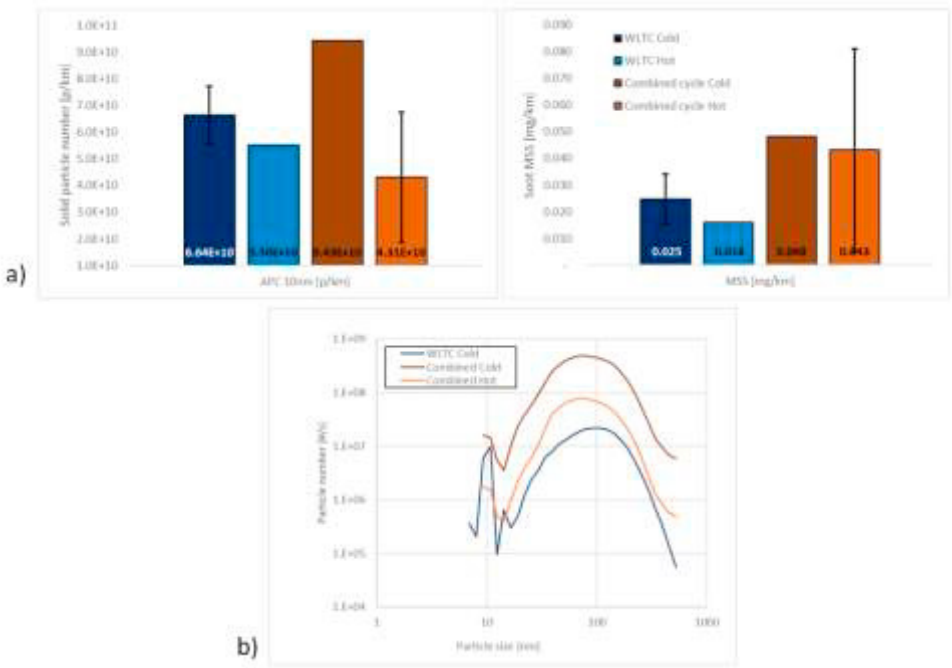

Fig. 6. (a) Average PN and soot emissions of the two cycles and (b) Particle size distribution on the two cycles.

4. Particle analysis

Particle analysis consists of chemical and toxicology analysis. The ALI system enables studies of the effects of the particles in situ, but the results will be compared with particle analysis using filters. The end filter in the DGI is made of Teflon, the pore size is 3 μm, and the nanoparticles will be collected, although the pore size is larger than the sub 100 nm particle emissions due to diffusion. The end filters from the DGI have to be cut into three pieces for PM physical, chemical and toxicological analyses. The pieces are sent to the three locations, for chemical analysis, for animal toxicology tests, and for submerged toxicology tests. The upper stages DGI filters are also used for further analysis. The whole set-up of filters from the ELPI+ is also analysed in terms of inorganic particle characterisation.

4.1. Particle physical and chemical characterisation

The sub 100 nm particle emissions generated in the field and lab are categorised, particularly for determining organic and inorganic chemicals and regarding size and morphology. Four nPETS partners do the handling of organic and inorganic chemicals. The chemical species that are included are dioxins, POPs, PAHs and metals, all relevant from a toxicological point of view. Preliminary tests for optimising the protocol for measurements were performed to assess analytical procedure performances. Before extraction, samples were spiked with a mixture of labelled standards. Extraction was carried out by Pressurized Liquid Extraction (PLE), using an accelerated solvent extraction instrument (ASE 300, Dionex, Sunnyvale, CA, USA) with dichloromethane, followed by a second extraction with acetone-dichloromethane for more polar chemicals.

The recovery results for PAHs can be divided between those corresponding to the most volatile compound from those with less volatile compounds. The first group recoveries (Np, Acy, Ace, F, Ph, and An) showed lower recoveries ranging from 35%±3% to 83%±2%, while for the reaming compounds, higher recoveries, ranging from 72%±3% to 95%±10%, were obtained. The recovery results for OPAHs ranged from 72%±2% to 101%±10%, while the recoveries percentage extraction rates varied from 89%±14% to 106%±14% for NPAHs.

Several elements were not directly assessable during the inorganic fraction chemical characterization of samples from exhaust emissions due to the blank composition, and qualitative results have been obtained. Nevertheless, thanks to a comparative analysis between samples and blank elemental compositions, it has been possible to identify specific markers for the fuels, i.e., elements which have been found in all the analysed nano-particulates: they are Phosphorous, Calcium and Zinc. In addition, other elements have been identified at least in some of the fuel's nano-particulates, such as Manganese, Iron, Chromium, Nickel and Copper. Fig. 7 reports an example of the performed measurement and a picture representative of the investigated samples. Further, the first map (not shown) of characteristic elements for the two different typologies of nano-particulates, i.e., from fuels exhaust and brakes non-exhaust, has been drawn.

A preliminary observation was performed using SEM to set the best conditions for morphological characterisation of nanoparticles. As a result, it was possible to appreciate some dispersed nanoparticles of 2-3 um diameter. However, it turned out that the grease substrate was not suitable for an optimal investigation. Thus, the grease substrate used for ELPI collection of nanoparticles does not represent the best solution to obtain a proper morphological characterisation.

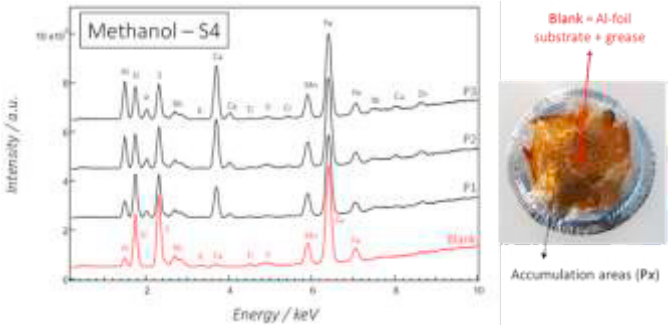

Fig. 7. Experimental XRF spectra were collected on samples from Methanol fuel on ELPI+ stage 4 Aluminum pre-greased substrate.

4.2. Particle toxicological analysis

We analyse particle toxicology in three main ways. Using the ALI system, particles on filters for submerged tests to study toxicological inflammatory markers, and exposing zebrafish embryos to particles.

Cell culturing and transportation and toxicity testing conditions have been verified for the ALI system. Also, optimal ALI conditions regarding temperature, high voltage, humidity, flow, CO2 concentration, particle deposition, cyclone and impactor cut-offs, and toxicity testing of in-lab particles. Initial tests in a metro station (see Fig. 3a-c), a road tunnel in Stockholm, and a spark discharge generator lab show promising results. However, the initial tests of particle toxicology in the subway and road tunnels stress the importance of measurement iterations.

The data generated from different filters and extraction methods have given some indicative results. For example, when using the EMFAB filter TX40, Ø70 mm. When the nanoparticles were removed in MilliQ and ethanol, low amounts of nanoparticles were removed from the filter, and it was a problem with a low mass of nanoparticles on the filter. Furthermore, a substantial part (approx. 30%) of extracted mass was due to fibers from the filter. No cytotoxicity was observed (THP-1 derived macrophages) from the particles or blank. Extraction using cell media was not feasible since this increased filter weight (and the mass of particles extracted is based on decreased filter weight). The PTFE (Teflon) filters were easier to handle, and extraction was possible without contamination with filter fibers. Approximately 50% of the mass of nanoparticles on the filters could be removed. No cytotoxicity from blank filter extract (MilliQ water and ethanol) was observed. Initial experiments on particles collected in a road tunnel showed no toxicity in doses tested up to 100 μg/mL. An increase in the release of the cytokine TNFα and IL-1β (only in MilliQ extract) was observed. MilliQ water seems to be the most promising approach.

5. Research collaborations

The nPETS projects are making use of the findings in earlier European projects having common partners with nPETSs partners, for example, nPETS makes use of DownToTen's sampling and dilution system for distinguishing fresh and secondary aerosols in lab. nPETS further nurse collaborations with the sister project ULTRHAS and the closely related project LEON-T and AURORA. Note that the results from the nPETS research will be collected in an nPETS database, including field and laboratory campaigns, toxicology analysis and particle characterization results, which will ease the exploitation of the findings.

Acknowledgements

This work is part of nPETS, a project that has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No 954377.

References

DownToTen, HORIZON 2020 Call: H2020 GV 2016 2017 Technologies for low emission light duty powertrains, Final Review Meeting, Brussels 28th February 2020.

Kerminen, V. M., et al., 2018. Environmental Research Letters, 13, 103003.

Latvala S., Hedberg J., Möller L., Odnevall Wallinder I., Karlsson H. L. and Elihn K., 2016. Optimisation of an air–liquid interface exposure system for assessing toxicity of airborne nanoparticles, J. Appl. Toxicol. 2016; 36: 1294–1301.

nPETS Grant Agreement, 954377, H2020-LC-MG-1-14-2020-TwoStages, Research & Innovation Action, Innovation and Networks Executive Agency, European Commission, 2021.

Rönkkö, T., Kuuluvainen, H., Karjalainen, P., Keskinen, J., Hillamo, R., Niemi, J.V., et al., 2017. Traffic is a major source of atmospheric nanocluster aerosol (Open Access) Proceedings of the National Academy of Sciences of the United States of America, 114 (29), pp. 7549-7554.

Simonen, P., Saukko, E., Karjalainen, P., Timonen, H., Bloss, M., Aakko-Saksa, P., Rönkkö, T., Keskinen, J., and Dal Maso, M., 2017. A new oxidation flow reactor for measuring secondary aerosol formation of rapidly changing emission sources, Atmos. Meas. Tech., 10, 1519-1537.

WHITE PAPER Ambient ultrafine particles: evidence for policy makers, 2019. Prepared by the 'Thinking outside the box' team, Coordinated by Flemming R. Cassee, Lidia Morawska, Annette Peters, the White paper is published on the European Federation of Clean Air and Environmental Protection Associations (EFCA) website: Ambient ultrafine Particles: evidence for policy makers.