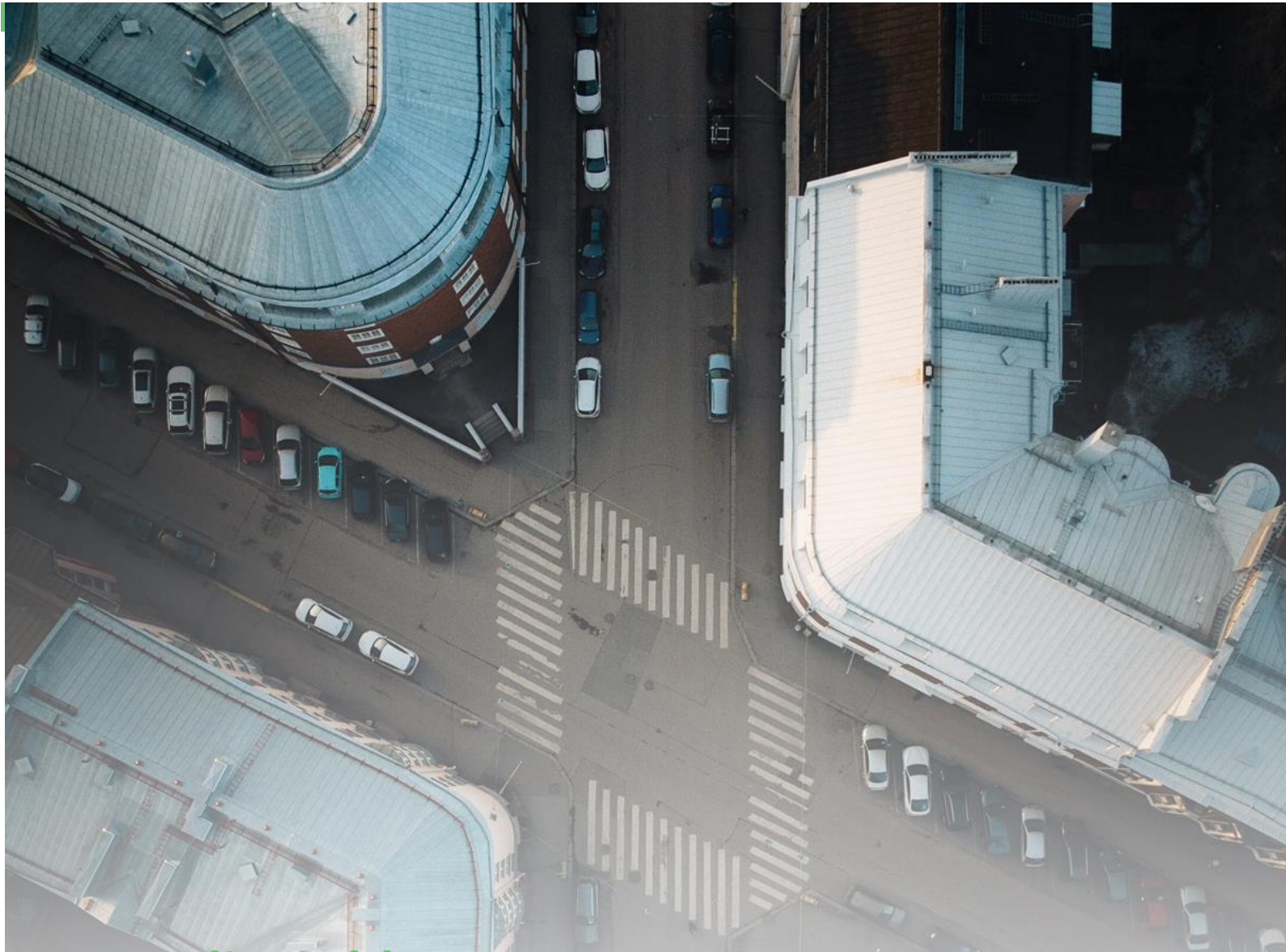

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PAREMPI
Particle emission prevention and impact: from real-world emissions of traffic to secondary PM of urban air

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Executive summary

This report summarizes the main findings of the *Particle emission prevention and impact: from real-world emissions of traffic to secondary PM of urban air (PAREMPI)* -project (https://parempi.eu/). Part I presents the scientific basis of secondary aerosol (SecA) formation from precursor emissions, and its impacts on health, air quality (especially in winter), and climate. Part II covers results of SecA quantification, toxicity studies as well as evaluation of external costs of SecA to society based on health impact assessment. An equivalent total particle emission index module (ePMI) is presented. Part III evaluates technologies to reduce harmful precursor emissions and mitigate SecA formation in EU urban areas. Robust measurement systems are suggested for measuring PM_{aged} (aged PM, including fresh and SecA emissions) emissions and toxicity from transport sources.

Traffic emissions consist of gaseous and particulate components originating from fuel, lubricating oil, and exhaust aftertreatment systems. Combustion produces large amounts of different SecA precursor compounds, which remain poorly characterized and quantified. Once released into the atmosphere, exhaust emissions cool, dilute and mix with ambient air pollutants, where gaseous precursor emissions can contribute to the formation of SecA (D3.1). In PAREMPI, understanding on secondary aerosol formation from precursor gases was evaluated and improved. Semi-empirical autoxidation mechanisms up to very condensable highly oxygenated organic molecules were developed for key single-ring aromatic hydrocarbons and modelling of alkane- and polycyclic aromatic hydrocarbon (PAH) functionalization progressed (D3.2). Modelling of aerosol dynamics remains challenging and current models simplify SecA formation.

In PAREMPI quantification of PM_{aged} from transport sources was approached by reviewing the state of knowledge, and by conducting complementary measurements. Four intensive measurement campaigns were designed to study emissions from road transport (cars, two trucks in winter conditions), marine (fuel sulfur, exhaust ammonia and ash as variables) and aviation (fossil and SAF fuels) sectors, and additionally brake dust emissions were studied (D2.2, D4.2). The results highlight the significance of SecA from transport sources with potential implications for both air quality and public health. The SecA formation was observed to depend on engine, fuel and aftertreatment leading to different contributions of organic and inorganic (SO_2, NO_x, NH_3) emissions. With cars and trucks, cold starts were identified to be important for SecA formation, as well as switching PHEVs to combustion engine operation. For modern vehicles emitting low levels of regulated emissions, the relative role of unregulated emissions, such as sub-10 nm particles and semi-volatile PAHs increased. PAREMPI experiments with marine and aviation fuels showed substantial fresh and PM_{aged} emissions. With distillate marine fuels, organics dominated PM_{aged}, while NH_3 in the exhaust caused a major shift towards nitrates and ammonium emission, and high-sulfur fuels towards sulfates, respectively. With aviation fuels, PM_{aged} was dominated by organics. The toxicity findings from the measurements suggest that even brief exposures can cause relevant toxicological responses, including emissions from the cleanest technologies, with ambient temperature having an important role. The break wear emissions depended on pad type and braking conditions. Moderate ultrafine particle emissions occurred during harsh braking. The SecA and toxicity results from PAREMPI measurements are collected in the database to complement existing data extracted from scientific literature (D5.1, D5.2). Still, several gaps in the knowledge remain: aviation and marine sectors lack in-field PM_{aged} measurements, and on-road measurements are scarce. Additionally, comparability and reproducibility across studies is limited due to non-standardized settings, calibrations, aging and protocols, and inconsistent reporting units.

PAREMPI also developes an ePMI module for estimating primary and secondary aerosol emissions from transport in global models (D3.4). The ePMI has been tested in ADCHEM and EC-Earth/TM5, and ongoing work focuses on integrating simplified anthropogenic SOA schemes analogous to

existing biogenic SOA schemes. In the modelling of vehicle emission impacts on the $PM_{2.5}$ and particle number concentrations in Europe shows that road transport significantly contributes to $PM_{2.5}$ (mainly nitrates) and ozone, especially in central Europe during late winter/early spring. The assessment of how vehicle emissions (road and ship traffic) impact the air quality, air pollution population exposure and population health (mortality and morbidity) in Europe was conducted. Mortality-related external costs were assessed, covering both direct (healthcare, social services) and indirect (productivity losses, informal care) impacts, noting that figures vary by country, healthcare system, and methodology. According to estimates, approx. 111,000 premature deaths per year in the EU-27 can be attributed to SecA exposure. The total annual societal cost of these health outcomes attributable to SecA in the EU-27 is estimated at €544 billion (D4.1).

In PAREMPI, technologies to prevent SecA formation were identified, for example, the results highlighted efficiency of tightening emission standards, although further improvements are needed, particularly during cold-starts and when PHEVs switch to combustion engine. From SecA precursor gases, attention is needed for NO_x, NH_3 and in marine and aviation sectors on SO_2 and monoaromatics content of exhaust. Amongst the SecA prevention technologies, robust measurement systems suggested by PAREMPI (**D2.3**), as well as ePMI tool, SecA and toxicity databases, and molecular-level modelling support policies to reduce SecA formation from transport sources. These aspects are assessed in policy recommendations report, PAREMPI D5.4.

Overall, PAREMPI advanced understanding of SecA formation and prevention methods through new data, modeling, as well as insights on toxicity and health effects. The ePMI software and SecA database enable improved $PM_{2.5}$ predictions and health impact assessments, supporting targeted mitigation and policy actions to reduce air pollution and health burdens. Further research is essential to fully understand the complex dynamics of SecA formation in real-world driving conditions. Knowledge gaps also remain on lack of standardized measurement protocols, portable instruments, and scarce data on SecA from aviation, marine and non-road sectors, and also from road transport in real-world conditions. Addressing these gaps is critical for risk assessment and regulations. Mitigation strategies include also systematic approaches, which will be addressed in PAREMPI policy recommendations (D5.4). It is clear that no single technology can prevent SecA formation across all transport sectors. Emission reductions generally follow improved energy-efficient engines, electrification, cleaner fuels, and advanced exhaust after-treatment.

List of abbreviations, definitions

ALI	Air-Liquid Interface
AK	Adenylate Kinase
BC	Black Carbon
BEAS-2B	Human Bronchial Epithelial Cell Line
C	Cytotoxicity (in toxicity rating)
CO	Carbon monoxide
CO_2	Carbon dioxide
CVS	Constant volume sampler
D	Different / Other Endpoints (in toxicity rating)
DEG	Differentially Expressed Gene
DMB	Diesel Marine Blend
DPF	Diesel Particulate Filter
EF	Emission factor
EFM	Exhaust flow meter
ePMI	Equivalent total particles emissions index module
FAME	Fatty acid methyl ester
FID	Flame ionization detector
FTIR	Fourier transform infrared spectroscopy
G	Genotoxicity (in toxicity rating)
H_2AX	Phosphorylated Histone H2AX (marker of DNA damage)
HD	Heavy duty
HIA	Health impact assessment
HOM	Highly oxidated organic molecules
HVO	Hydrotreated Vegetable Oil
LD	Light duty
NH_3	Ammonia
Nitro-PAH, NPAH	Nitrated PAHs
NO_x	Nitrogen oxides
nv	Non-volatile
OC	Organic Carbon
OF	Oxidation Flow
OFR	Oxidation flow reactor
OH	Hydroxide
Org	Sum of organic compounds (mass)
O	Oxidative Damage (in toxicity rating)
OVOC	Oxygenated VOCs
Oxy-PAHs, OPAHs	Oxygenated PAHs
PAH	Polycyclic aromatic hydrocarbons
PAM	Potential aerosol mass chamber
PEMS	Portable Emissions Measurement System
PM	Particulate matter (mass)
PM_{aged}	Particulate matter (mass), aged particles including SecA
PM_{fresh}	Particulate matter (mass), fresh particles before ageing
PM_{fresh} (reg)	Fresh PM measured with regulated method
PM_{fresh} (AMS)	Fresh PM measured with AMS (size class approx. 50-700nm)
PM_{fresh} (ELPI)	Fresh PM measured with ELPI

PM_{aged} (AMS)	Aged PM measured with AMS
PM_{aged} (ELPI)	Aged PM measured with ELPI
PN	Particle Number
PN23	Number of particles larger or equal to 23 nm in diameter
PN10	Number of particles larger or equal to 10 nm in diameter
$PN_{tot}2.3$	total PN with Dp >2.3 nm
$PN_{nv}23$	Non-volatile PN with Dp >23nm
$PN_{totAged}$	Aged total PN concentration
PTR-MS	Proton Transfer Reaction Mass Spectrometer
RH	Relative humidity
RNA-seq	RNA Sequencing
ROS	Reactive Oxygen Species
RDE	Real Driving Emissions
SecA	Secondary aerosol, includes SOA+SIA
SIA	Secondary inorganic aerosol
SOA	Secondary organic aerosol
SNR	Signal-to-noise ratio
SoC	State of Charge of the battery
STP	Standard temperature and pressure, 273.15 K, 100 kPa
THC	Total hydrocarbons
T	Timepoint (e.g. T0, T24, T48)
TEER	Transepithelial Electrical Resistance
UFP	Ultrafine Particles, particles with diameter <100nm
UV	Ultraviolet
VOC	Volatile Organic Compounds. Sub-classes: anthropogenic (AVOC), biogenic (BVOC), oxidated (OVOC), highly oxidated (HOM)
VBS	Volatility Basis Set

Definition of terms used in PAREMPI

Currently, multiple overlapping terms are used to describe various particulate and gaseous components in exhaust emissions, which can lead to confusion and inconsistencies. To ensure clarity and consistency across all PAREMPI outputs—and to support future use of the data—a standardized set of terms and definitions has been established for commonly measured particle phase exhaust components. For both particulate mass (PM) and particle number (PN), the naming convention follows a structured format including following parts:

Primary identifier:

- The measured component (e.g., PM or PN).
  - PM: Emitted PM consists mainly of black carbon (BC), organics, sulfates, nitrates, and ammonium.
  - PN: the particle size cut-off applied during collection is specified; for example, $PN_{tot}2.3$ denotes the total PN with D_p >2.3 nm. The abbreviation for total particles is tot, for example, $PN_{tot}23$.
  - Black carbon (BC): In aviation sector, measured BC is called “non-volatile PM”.

Condition:

- Fresh: Refers to diluted and cooled exhaust that does not include secondary aerosol (SecA). Fresh particles may contain condensables, depending on dilution system and its conditions used in the measurements.
- Aged: Refers to diluted, cooled, and aged exhaust, which includes both the fresh emissions and the SecA formed through atmospheric oxidation (mimicked e.g. via an oxidation flow reactor, OFR). For example, PM_{aged} denotes the total particulate mass measured after the OFR, encompassing both PM_{fresh} and SecA. SecA consists of secondary organic aerosol (SOA) and secondary inorganic aerosol (SIA). PM_{aged} emissions can be measured by combining BC and non-BC composition data from AMS, in addition to conversions from size-distributed particle number data.
- Non-volatile PN: Non-volatile (nv) particle number emissions are measured after the thermal treatment of the fresh exhaust aerosol. For example, $PN_{nv}23$.

Measurement method:

- If measured using a regulated (reg) method, this is indicated.
- If not, the instrument used is specified, for example PM_{fresh}(AMS), PM_{fresh}(ELPI), PM_{aged}(AMS+BC), PM_{aged}(ELPI)

Organic and inorganic subfractions (POA, CPOA, semi-volatiles, SOA, SIA)

The organic fraction of particulate matter (PM) comprises tens of thousands of different compounds. To better understand and characterize this complexity, various subgroups are defined based on formation processes, chemical composition, or emission sources. Two commonly used terms include **Primary Organic Aerosols (POA)** for directly emitted particles from sources such as combustion and **Secondary Organic Aerosols (SOA)** for particle mass formed in the atmosphere through chemical reactions of volatile organic compounds (VOC). These definitions are further detailed in Section 1.4 and the **PAREMPI D3.1** report on aerosol dynamics during exhaust dilution and cooling. A specific subset of POA is **Condensable Primary Organic Aerosols (CPOA)**, which include semi-volatile compounds. These substances exist in the gas phase in hot, undiluted exhaust but can condense into particles within seconds to minutes as the exhaust cools in the atmosphere. The quantification of CPOA emissions is highly dependent on the sampling system used, making absolute emission estimates uncertain. In regulated PM sampling strict control of dilution ratio, temperature, face velocity, and other parameters enable repeatable measurements, even when PM contains CPOA. Notably, PM can contain CPOA even before it exits the tailpipe. For instance, when measured inside the exhaust duct, the presence of CPOA in PM depends on the specific application and fuels used. This is particularly relevant for diesel engines operating on conventional marine fuels (Aakko-Saksa et al., 2018). In addition to organic aerosols, **Secondary Inorganic Aerosols (SIA)** are formed from inorganic precursor gases through atmospheric reactions. SIA includes sulfate, nitrate, ammonium and chloride emissions. When PM includes both SOA and SIA, the term **SecA** (Secondary Aerosol) is used. This harmonized terminology improves data transparency and comparability, enabling more effective integration and interpretation of results across different studies and transport sectors.

PART I: Scientific basis on the secondary aerosol formation from transport emissions

1 Understanding of transport emissions as SecA precursors

1.1 Formation and atmospheric transformation of exhaust emissions

The combustion of hydrocarbonaceous fuels in the presence of oxygen ideally produces only carbon dioxide (CO_2) and water. However, real-world combustion is non-ideal and results in a small fraction of constituents, referred to as “emissions”, typically comprising less than 0.5 vol% of the exhaust flow. Spark-ignited (SI) engines operating near the stoichiometric air-to-fuel ratio (AFR~1) primarily emit CO_2, H_2O and N_2, while diesel engines operate with excess air (lean combustion, AFR > 1) and therefore also emit oxygen. Combustion in aircraft turbines is inherently lean and operates with high excess air. Besides the major exhaust components, air-polluting exhaust emissions include organic gases (OG), carbon monoxide (CO), nitrogen oxides (NO_x), ammonia (NH_3), nitrous oxide (N_2O), and particulate matter (PM) depending on the engine, fuel and exhaust aftertreatment technology (EAT) used. PM emission consists of black/elemental carbon (BC/EC), organic compounds, inorganic ions (such as sulfates, nitrates, ammonium) and trace metals. Organic gases encompass compounds such as hydrocarbons (e.g. methane, benzene) and carbonyls (e.g. formaldehyde, acetaldehyde). Many organic compounds, including polyaromatic hydrocarbons (PAHs), nitro-PAHs, and oxy-PAHs, are present in both the gaseous and PM phases. A significant number of these compounds are semi-volatile with dynamic gas-particle partitioning depending on the prevailing conditions.

Engine-out emissions depend on both fuel type and engine technology. In diesel combustion, engine-out NO_x and PM emissions are typically high, while OG and CO emissions remain relatively low. Alternative fuels can be used in to support cleaner combustion; however, these fuels may also lead to new emission species, such as unburned methane when engines are fuelled with natural gas or biomethane. Substantial emission reductions can be achieved by EAT systems. Engines operating near stoichiometric AFR can utilize three-way catalysts (TWC) to reduce CO, HC, and NO_x emissions. To achieve low NO_x and PM emissions in diesel engines, EAT systems, such as selective catalytic reduction (SCR) and diesel particulate filters (DPF), are required. Some emission species are formed in EAT systems. For example, NH_3 and N_2O can be formed as by-products of catalytic reactions in a TWC. Urea-based SCR systems can slip NH_3 emissions under conditions of urea over-dosing or incomplete conversion.

Ambient traffic emissions consist of a complex mixture of gaseous and particulate components originating from fuel combustion, lubricating oil, and EAT systems, as well as non-exhaust emission sources such as brake and tire wear. Once released into the atmosphere, exhaust emissions cool, dilute and mix with ambient air pollutants. Gaseous emissions can participate in atmospheric chemical reactions and condense into the particulate phase, contributing to the formation of SecA (e.g. Rönkkö and Timonen, 2019). This transformation is complex, involving interactions between exhaust species, such as SO_2, NO_x, NH_3, and VOCs, and ambient atmospheric compounds. Overall, ambient $PM_{2.5}$ in urban environments accumulates from many sources, including also other anthropogenic and natural sources, as well as long-range transported pollutants. Consequently, the

source apportionment of $PM_{2.5}$ and the assessment of its health and environmental impacts remain challenging. Atmospheric pollutants from transportation adversely affect air quality, human health, and climate, leading to substantial societal costs. The pathway from emissions to ambient PM and its impacts, including atmospheric transformation processes, are illustrated in Figure 1.

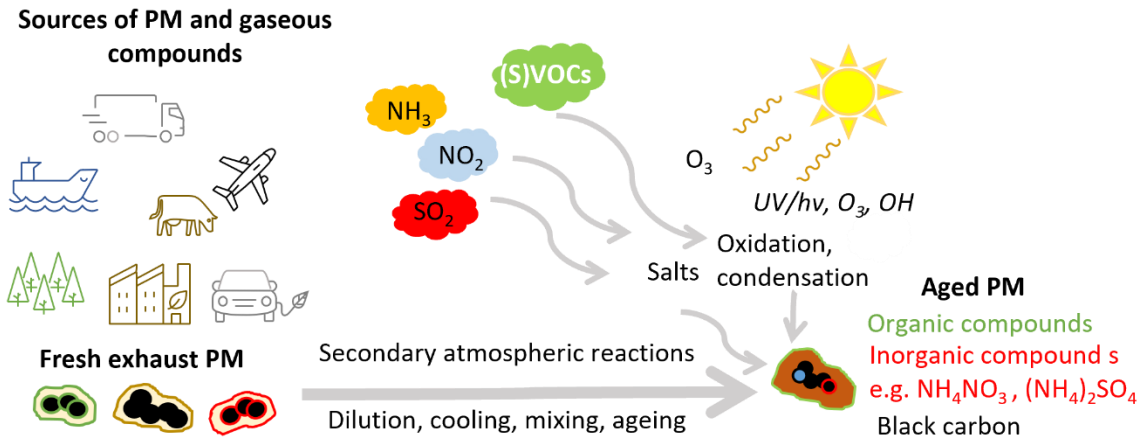


Figure 1. Schematic presentation of the growth of fresh PM to aged, ambient PM through atmospheric reactions during dilution, cooling, and aging (Adapted from Aakko-Saksa et al., TRA2024).

Recent studies (e.g. Timonen et al., 2017, Kuittinen et al., 2021) have indicated that SecA mass formed from gaseous precursors can be significantly higher than the mass of particulate matter in the exhaust, i.e. regulated PM emission. A recent study by Pye et al. (2021) found that secondary organic aerosol is strongly associated with county-level cardiorespiratory death rates in the U.S., independent of the total $PM_{2.5}$ mass association. Despite all advances and the importance of the topic, our understanding of the SecA formation process remains limited (Zhang et al., 2024). This is especially true for different fuels, engine technologies, and aftertreatment systems, particularly under winter conditions. For example, air quality is worse at low winter temperatures with low atmospheric mixing height, however, dependences causing this phenomenon, and particularly the role of transport sectors' emissions, are not evidenced. Moreover, the role of SecA in particle toxicity and its broader impacts on air quality, health, and climate are poorly constrained.

1.2 SecA precursor emissions (VOCs, SO_2, NO_x, NH_3)

The following paragraphs discuss organic and inorganic transport-related emissions that act as precursors to SecA formation. These include organic gases and inorganic precursors such as sulfur oxides, nitrogen oxides, and ammonia (Renner and Wolke, 2010). In the atmosphere, organic gases may react with OH radicals and ozone (O_3) during the daytime or with O_3 and NO_3 radicals at the nighttime (Srivastava et al., 2022).

Emissions of organic gases

In the atmosphere, thousands of different volatile organic compounds (VOCs), semi-volatile (SVOCs) and intermediate-volatility organic compounds, mainly biogenic VOCs (such as isoprene and terpenes), are present (Srivastava et al., 2022). Transport applications emit hundreds of individual organic compounds in engine-out exhaust, including various types of alkanes, alkenes, cycloalkanes, cycloalkenes, aromatic compounds, carbonyls and nitrated organics. Among these, single-ring aromatics and substituted derivatives, such as benzene, toluene, ethylbenzene and xylene (BTEX), are evidenced or suspected precursors of SOA formation (see e.g. Suarez-Bertoa et al., 2015, Timonen et al., 2017). Characterization of SOA formation potential of individual organic compounds is challenging, as discussed in section 1.3.

Aromatics in the exhaust may originate from either the fuel or lubricating oil. Fossil fuels typically contain some aromatic species, but fuels can be also aromatic-free depending on the production processes and requirements for the final fuel. As regards lubricating oils, feedstocks used to produce mineral base oils contain aromatics, but those are removed during solvent extraction and hydroprocessing (Whitby, 2018). Even if hydrocarbons do not enter combustion as aromatic compounds, the formation of aromatics is possible during the combustion process. Aliphatic precursors can be for example sufficiently large alkanes or alkenes (e.g. C_5-C_{17}), which are volatile enough to stay in the gaseous phase prior to oxidation. Large hydrocarbons may be released into exhaust also from incompletely burned fuel and lubricating oil (Carbone et al., 2019).

Semi-volatile organic compounds (SVOCs) may be emitted from transport applications, originating from both fuel and lubricating oil. However, these compounds are typically not included in emission inventories, which mainly account for light VOCs with fewer than 12 carbon atoms, such as the single-ring aromatics mentioned earlier. SVOCs are generally heavier; for example, the US Environmental Protection Agency (EPA) defines them as compounds containing more than 12 carbon atoms and having boiling points between 240-400 °C (US EPA 40 CFR 1065.1103–1111). Emission factors for the semi-volatile fraction of exhaust from transport sources are scarce, although individual compounds, such as certain polycyclic aromatic hydrocarbons (PAHs), have been reported. Lu et al. (2018) reported of gas-phase and particle-phase profiles for on-road and off-road gasoline, gas-turbine, and diesel engines. Measuring SVOCs requires special sorbents and analytical methods, and quantifying their total mass is not straightforward. Due to their dynamic gas-particle partitioning, SVOCs may be present in either the gas or particulate phase depending on the measurement conditions, such as temperature and pressure. Omitting SVOCs from atmospheric modelling can lead to an underestimation of atmospheric SOA formation from transport sources (Lu et al., 2018). Moreover, VOC and SVOC emissions tend to increase at low ambient temperatures due to prolonged engine and EAT warm-up periods. As a result, aromatic emissions may be elevated during the winter season.

As previously mentioned, unburned fuel is a source of organic emissions. Therefore, fuel composition, when considered alongside grouped emissions, can indicate expected aromatic emissions in VOC and SVOC phases. In the results of the PAREMPI measurements (see section 3), non-methane organic gases (NMOG) are presented to indicate the potential presence of organic precursors for SOA formation in exhaust gases. However, a comparative analysis between fuel composition and measured aromatic emissions remains pending and will be addressed in forthcoming publications.

Sulfur oxide emissions

Most of the sulfur present in exhaust gases, originating from fuel and lubricating oil, is oxidized during combustion in internal combustion engines to form SO_2. A smaller fraction is emitted as sulfur trioxide (SO_3) and other sulfur-containing compounds. Reportedly, SO_3 typically accounts for approximately 1-2% of the SO_x formed from fuel sulfur. SO_3 can further react to form sulfuric acid and sulphates, which contribute to the engine-out PM emissions. The conversion of SO_2 to SO_3 depends on several factors, including engine load, air-to-fuel ratio, combustion temperature, and the presence of vanadium (in fuel or SCR), which can enhance the conversion rate. In engine-out PM, the proportion of hydrated sulfate species can vary significantly depending on the fuel type and EAT. For example, it may range between 22%–62% for high-sulfur marine fuel, while being around 8.3% for distillate marine fuel. (Grigoriadis et al., 2021, Stuart, 2010).

Modern vehicles are typically equipped with exhaust aftertreatment systems that aim to oxidize exhaust compounds, which can significantly increase also the oxidation of sulfur compounds, leading to increased formation of sulfur trioxide and further, formation of gaseous sulfuric acid in the hot and undiluted exhaust. Due to the temperature dependence of oxidation efficiencies of exhaust

aftertreatment systems, also the exhaust sulfuric acid concentration can have strong dependence on exhaust temperature, or on the other hand, on the driving situations; higher sulfuric acid concentration is observed at higher engine loads when also the exhaust temperature is at high level. However, exhaust aftertreatment systems can also act as storage for sulfur compounds, and vice versa, release of sulfur compounds to exhaust flow from these storages. Exhaust and traffic environment studies indicate that exhaust sulfuric acid can contribute significantly to the formation of new particles when the exhaust is cooled and diluted when emitted to the atmosphere. This particle formation likely occurs through nucleation processes that also involve other compounds, such as oxidized organic molecules. As a result, the total number of particles can increase, and a distinct *nucleation mode* can appear in the particle size distribution of fresh exhaust (e.g. Olin et al., 2020).

Nitrogen oxide emissions

Nitrogen oxides are formed in high temperature combustion through the reaction of nitrogen and oxygen in intake air. Since the nitrogen content of transport fuels in transport sectors is low, it does not significantly contribute to NO_x formation, except in potential future applications involving ammonia as marine fuel. Engine-out NO_x emissions are particularly high in diesel engines but can also be substantial in stoichiometric SI engines. NO_x emissions mainly consist of NO, with a smaller fraction of NO_2 in diesel combustion.

Ammonia emissions

Atmospheric ammonia originates mainly from agricultural sources. NH_3 emissions from transport applications are formed mainly in the EAT systems. Ammonia can enter the exhaust from selective catalytic reduction (SCR) systems from excessive ammonia or urea injections (Zhang et al., 2022). Ammonia can also be formed from side reactions in a three-way catalytic (TWC) converter when NO reacts with CO and H_2O or H_2 (Mejía-centeno et al., 2005, Suarez-Bertoa et al., 2016, Jiang et al., 2023). Additionally, future marine engine applications may utilize ammonia as a fuel and emit NH_3 also as unburned fuel. Simultaneous emissions of NH_3 and PM may occur under certain operating conditions, contributing to complex exhaust compositions and potential secondary aerosol formation (Simon et al., 2025).

1.3 Secondary organic aerosol formation from representative compounds (at molecular level)

Several of the gaseous components emitted by traffic have a high secondary aerosol formation potential. The strong acids and bases (e.g., sulfuric and nitric acids, amines and ammonia, respectively) are largely responsible for the stable initial clusters that work as seed particles for the subsequent secondary material growth (Olenius and Roldin, 2022). This further secondary material is most readily provided by a multitude of oxidized organic compounds that are generally the most abundant material present capable of influencing early particle growth (Tröstl et al., 2016). The condensable material is formed by rapid autoxidation processes that transform the gaseous precursor molecules into practically non-volatile aerosol embryos even in sub second timescales (Bianchi et al., 2019, Iyer et al., 2021). The condensation of these oxygenated organics results in the formation of SOA, the most abundant form of ambient submicron aerosol mass and the likely culprit in several toxicological effects. In the case of traffic, the most common sources of SOA are aromatic and polyaromatic compounds, followed by large alkanes and alkenes.

Traffic emitted VOCs transform into SOA in the ambient gas-phase through complex oxidation networks. Often the most facile paths to low-volatile aerosol forming material are via autoxidation chain reactions (Rissanen et al., 2014). During autoxidation the molecule rapidly increases its

oxygen content, and hence also weight, by repeated radical intermediate isomerization and subsequent O_2 addition steps, leading to direct aerosol precursors termed highly oxygenated organic compounds (HOMs) (Bianchi et al., 2019). Autoxidation is a particularly efficient path of molecular functionalization in the case of cyclic VOCs such as the traffic related single ring aromatics and polyaromatics (Iyer et al., 2023, Garmash et al., 2020). In these processes, the carbon-ring-breaking leads to reactive intermediates that are ideal at autoxidizing and rapidly transform the non-polar gaseous VOCs into highly polar and practically non-volatile aerosol embryos with high affinity for surfaces and liquid water. Autoxidation plummets the organic compound vapor pressure by each successive oxygen addition, and according to recent research HOMs are almost perfect molecular building blocks for subsequent SOA formation (Ehn et al., 2014).

To allow characterizing external and internal influences on early steps of SOA formation, e.g., change of clustering and condensing compound concentrations and change in chemical composition of the resulting SOA particle, a molecular level approach appears necessary. Unfortunately, the number of participating compounds increases exponentially during sequential oxidation generations, and consequently a full description of the oxidation process for each VOC is beyond any realistic vision. Thereby the grand aim of the current SOA modelling approach is to enable practical molecular level modelling of any traffic related VOCs with the help of prototypical reaction mechanisms constrained by laboratory flow reactor experiments of autoxidation product distributions as a function of reaction time. With this procedure, semi-empirical autoxidation networks up to the highly condensable HOM products can be developed for any VOC of interest. Here of special interest are traffic related single-ring aromatic hydrocarbons, but also polyaromatics (Iyer et al., 2023). The mechanistic steps propagating simple alkane autoxidation are generally simpler due to the saturated nature of the participating compounds, leading to far less competing reaction pathways.

Accurate molecular level SOA modelling is severely complicated by the missing rates and branching in crucial autoxidation pathways. Thereby, the alternative semi-empirical modelling approach applied in PAREMPI is attempting to alleviate the problem by bringing the best from both worlds: the best prior experimental understanding of the reaction mechanism (*i.e.*, the master chemical mechanism, MCM; Jenkin et al., 1997) combined with high-level quantum chemical computations of the critical reaction steps (e.g., Iyer et al., 2021; Iyer et al., 2023), to build a base model. As autoxidation, the critical component generating SOA forming vapors, is currently missing from all readily available chemical models, we simulated the autoxidation chemistry utilizing our recently developed semi-automated peroxy radical autoxidation mechanism code (*i.e.*, automated alkoxy/peroxy radical autoxidation mechanism framework “autoAPRAM-fw”, Pichelstorfer et al., 2024; Roldin et al., 2019). It uses laboratory measured oxidation product distributions, specifically including the least volatile reaction products currently missing from atmospheric chemical models, to assign rates and branching for the rest of the individual reaction steps. The result is a time-dependent molecular reaction product distribution as a function of starting conditions that can be used for the subsequent aerosol modelling.

A set of benchmarking environmental chamber SOA experiments with three Xylene (C_8H_{10}) isomers that differ only by the positions of the methyl groups were set up to critically assess the performance of the performed **molecular level modelling.** While the benchmarking experiments agree over wide timescales and experimental conditions, challenges remain in modelling the resulting molecular level SOA mass formation. Whereas the modelling does return most of the SOA mass formed in the isomeric Xylene chamber oxidation, and the measured mass does show the correct trends together with the deduced reaction mechanism, it is currently unclear where the biggest uncertainties remain. A rough schematic of the modelling framework and the uncertainty propagation are illustrated in Figure 2.

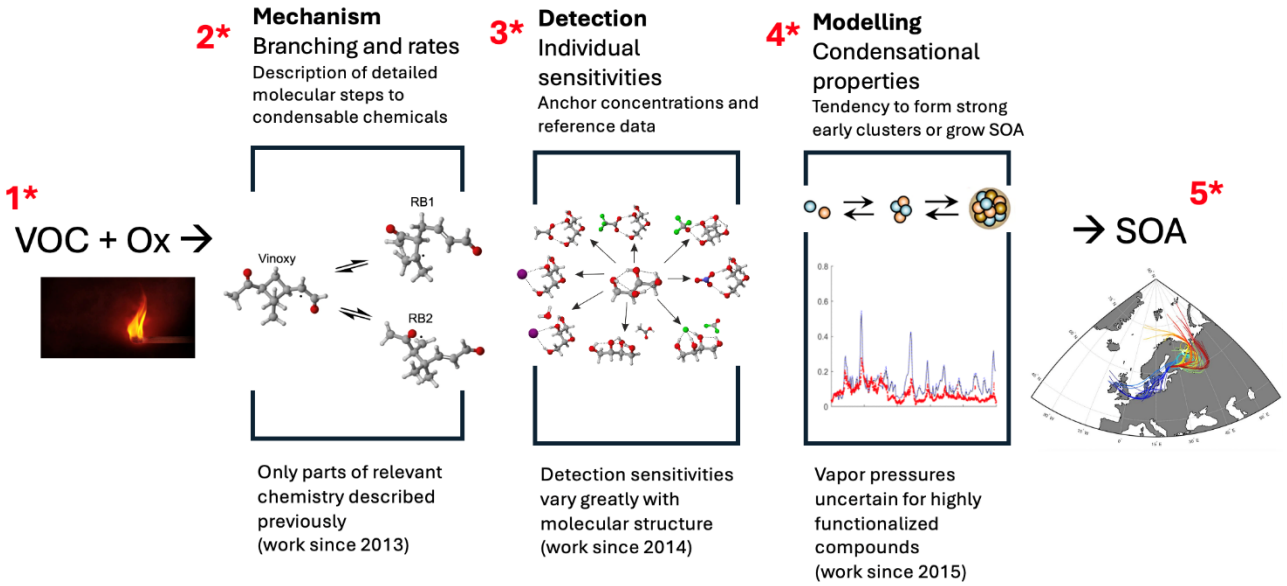


Figure 2. Benchmarking the molecular level modelling currently halted by poorly constrained uncertainties. The uncertainties propagate through the modular modelling process and consequently can result in large uncertainties in the final SOA modelling. Currently it is unclear how much the different uncertainty sources contribute to the overall uncertainty.

The overall performance is hinging on five major uncertainty sources, each contributing a considerable yet uncertain factor to total uncertainty. Factor 1 is related to the primary oxidation initiation rate, which can vary widely among different VOCs but can often be estimated from similar compounds with acceptable precision within a factor of five or so. Factor 2 originates from uncertain branching and rates between the important oxidation propagating and terminating channels. Due to the unknown nature of the unknown pathways, it is very hard to assign any realistic uncertainty range. Factor 3 relates to using chemical ionization detection, with widely varying detection sensitivities between structurally different molecular products. Factor 4 originates from computing and estimating the required molecular characteristics needed for subsequent SecA modelling. Vapor pressure is the key for predicting condensational properties, but for highly functionalized molecules the values are necessarily uncertain due to poorly constrained influence of internal degrees of freedom on oxygenated product vapor pressure. Thus, a combination of approximative methods must be utilized. Factor 5 relates to the ultimate secondary aerosol modelling and the related aerosol dynamics in various environmental conditions during secondary aerosol growth. Also, we note that in real-world conditions (in the atmosphere) there are also large uncertainties related to the emissions of the precursors (emission factors of VOCs) and we don't have good observations of the temporal and spatial variation of VOC concentrations or OH in the atmosphere.

Due to the well-recognized complex uncertainties in resolving unique molecule specific parameters, the PAREMPI project selected a semi-empirical modelling of the autoxidation networks that result in secondary aerosol formation as the method of choice for ambient aerosol modelling. This approach avoids the need for tedious molecule specific parameters for each oxidation product, and instead uses several experimental constraints (i.e., the product formations as a function of time) from the laboratory. It should be emphasized that it is not a trivial task to track and quantify the remaining individual uncertainties due to their dependent and tangled nature, and thus the semi-empirical approach is likely the most viable avenue also at least for the near future before automated quantum chemistry based routines and computing power mature to the level where they are able to provide complex molecular level predictions. Hence, a series of sensitivity analyses utilizing neural networks are planned to pinpoint the most crucial factors preventing ultimate molecular level closure.

1.4 Atmospheric fresh and aged aerosol formation, especially in winter conditions

The formation of submicron aerosol particles generally involves a physical phase transition from gas to condensed phase (via condensation or chemical reactions at particle surfaces); for soot it occurs in the flame, for primary organic aerosols (POA) in the cooling flue gases and for secondary organic and inorganic aerosols (SOA, SIA) in the atmosphere. Soot particles are formed during high temperature combustion of carbonaceous fuels in fuel rich regions, when polycyclic aromatic hydrocarbon (PAH) subunits are clustered into three-dimensional structures of 1-2 nm in diameter. During the formation process, these individual particles coagulate and coalescence together to form nearly spherical primary soot particles of around 15-20 nm. Finally, the primary particles coagulate and form chain-like fluffy agglomerate structures that are emitted into the atmosphere e.g. from road vehicles. PAH that does not convert to soot in the combustion process may instead be emitted to ambient air. This is important since several PAH molecules are genotoxic (for example B(a)P).

Typically organic aerosols are divided into primary and secondary compounds. Primary organic aerosols are traditionally defined as emitted directly (or freshly) into the atmosphere as aerosol particles. However, for the majority of POA, which are generated by combustion processes, this assumption may not be valid at the urban street canyon scale. The ambient temperature will also influence the new particle formation and condensation and evaporation dynamics during the exhaust dilution and cooling phase. When the flue gases cool down the inorganic (NH_3, H_2SO_4) and organic condensable compounds, including PAHs, can either condense on existing soot particle surfaces or contribute to the formation of new sub-10 nm particles (Karjalainen et al., 2014; PAREMPI Deliverable **D3.1**). Secondary organic aerosols are formed in atmosphere when biogenic and anthropogenic volatile organic compounds (BVOC, AVOC) are oxidized by oxidants, such as O_3, OH and NO_3 radicals and form condensable oxidation products (OVOC). Only a fraction of the formed OVOC will be low volatile enough to contribute to the SOA formation. A great challenge when it comes to the modelling of SOA is to represent how the OVOC composition and volatility distribution changes depending on the VOC mixture, the oxidants (O_3, OH, NO_3) and NO_x levels, and the temperature in the atmosphere.

Temperature and photochemistry impacts on the UFP formation. The physical phase transition from gas to condensed phase via condensation or nucleation, either in the hot exhausts or the ambient atmosphere depend on the vapour concentrations and their volatility (saturation vapour pressures) and the evaporation rates of the formed molecular clusters/particles. Both the nucleation and condensation processes and the formation of HOM that govern the formation and growth of UFP in vehicle exhausts and during atmospheric new particle formation events are strongly temperature dependent processes. At too high temperatures the forming molecular clusters will evaporate before they reach critical sizes where they are stable enough to grow into new aerosol particles by condensation. Simultaneously many of the potentially condensable vapors will be too volatile to contribute to the growth of nanometre sized particles. Hence, the ambient temperature will affect both the UFP formation in the cooling and diluting exhausts that leave the tailpipe, the condensable, semi-volatile, primary organic aerosol gas-particle equilibrium and the subsequent formation of SecA in the atmosphere. In general, low temperatures (wintertime conditions) will favour the fresh exhaust UFP and OA formation in the vehicle exhausts, as has been indicated in a previous experimental studies (e.g. Saarikoski et al., 2024) and demonstrated by our detailed aerosol dynamics model simulations presented in the PAREMPI **D3.1** report.

Secondly, the ambient SecA formation depend both on the photochemistry (sunlight intensity) and the atmospheric temperatures. Atmospheric new particle formation is driven by the formation of strong acids, in particular sulfuric acid (H_2SO_4), which is formed in the gas-phase when

SO_2 is oxidized by OH radicals. OH radical is primarily formed when O_3 is photolyzed by UV-light, which is favourable during the spring and summer in Europe. Thus, atmospheric new particle formation is a process which largely is driven by photochemistry and is hence supressed during the dark winter months but favours by days with high UV-light intensity but still low enough temperatures for the forming molecular clusters (e.g. H_2SO_4-NH_3) to be stable enough to growth into new nanometre sized aerosol particles. This likely explains why atmospheric NPF events are frequently observed during the spring in Europe. Nitric acid (HNO_3) is analogous to H_2SO_4 formed in the gas-phase when NO_2 is oxidized by OH. HNO_3 is a strong acid, but weaker than H_2SO_4, and the partitioning of HNO_3 to the particle phase requires some base that neutralizes the acidic aerosol aqueous phase. This base is typically NH_3. The gas-to-particle conversion and formation of ammonium nitrate (NH_4NO_3) PM is also favoured by low temperatures and humid conditions. These conditions are generally met during early spring in Europe (Bressi et al., 2021; Atabakhsh et al., 2025). However, the NO_2 + OH reaction is ~15 times faster than SO_2 + OH and the nitrate PM can also form during the night by heterogenous reactions involving N_2O_5. As an example, Eriksson et al. (2017) showed that NH_4NO_3 PM rapidly coat and transform diesel soot particles from initially hydrophobic agglomerates into nearly spherical and hygroscopic particles during dark and humid winter conditions in and downwind Copenhagen.

The atmospheric oxidation of the major anthropogenic SOA precursors (aromatic hydrocarbons and alkanes) is mainly initiated by OH radicals. Hence, ASOA formation is favoured in the spring and summertime when the UV-light intensity is high. Recent experimental studies have shown that aromatic compounds and C6–C10 alkanes can produce considerable amounts of highly oxygenated organic molecules (Iyer et al., 2023; Wang et al., 2021), and since the peroxy radical autoxidation reactions that produce HOM is highly temperature dependent this will likely further favor the ASOA formation in the summertime and at least partly compensate for the increasing SOA volatility at higher temperatures. Thus, at high photochemistry and high temperature conditions, typical during the summertime in Europe, we can expect that the OA generally should be composed of a larger share of low-volatility highly oxygenated SecA than during the wintertime when the OA is likely to be more dominated by less oxygenated primary OA. These trends will also be promoted by the strong and opposite seasonal trends in the BVOC emissions and the primary OA emissions from residential heating in Europe.

The temperature dependent formation of SecA needs to be considered more realistically by atmospheric models. Simulations using global CTMs, with simplified semi-empirical (no temperature dependence) NPF parameterizations suggest that NPF is the dominant source of the total aerosol particle number concentration, and an important contributor to the CCN budget on a global scale (Makkonen et al., 2012; Dunne et al., 2016). However, the actual contribution of NPF to the aerosol particle budget and the CCN concentrations in different environments are poorly quantified, which seriously hinders our ability to estimate the climate and health effects of atmospheric NPF, and especially the impact from anthropogenic sources (Kerminen et al., 2018). This fundamental knowledge is also a prerequisite to understand how emissions and atmospheric chemistry influence the size resolved SecA concentrations in urban and rural environments and to assess their impact on the properties of clouds, the radiation balance, air quality and human health.

In atmospheric aerosol dynamics models, SecA formation is represented by three distinct processes: nucleation, condensation, and coagulation. The formation of new aerosol particles via molecule clustering (nucleation) are generally parameterized by a NPF formation rate (J) for particles with a diameter of 1-2 nm. However, fundamentally at the molecule cluster level, these three processes cannot be distinctly separated and can therefore not be fully represented by simplified J-parameterizations. They are highly intertwined gas-molecular cluster-aerosol dynamic processes, which influence the complete size spectrum from single molecules and molecular clusters to large aerosols and clouds droplets. This also poses computational challenges and limitations on how

realistically secondary aerosol particle formation can be represented in global scale CTMs and ESMs (Olenius and Roldin, 2022). Due to computational constraints, the gas-phase chemistry mechanisms and SOA parameterizations are to a large extent decoupled in global CTMs and ESMs. The organic condensable products are generally represented by a few lumped volatility distribution species classes, a Volatility Basis Set (VBS; Schervish and Donahue, 2020). These VBS parameterizations are generally derived from laboratory experiments performed at specific and far from atmospheric realistic conditions (i.e. very different RO_2 bimolecular vs unimolecular reaction fate distributions) (Kenagy et al., 2024). Hence, these parameterizations will not be able to fully capture the temperature dependent and complex nonlinear RO_2 chemistry that governs the formation of OVOCs and SOA in the atmosphere. An important example is the interaction between NO, NO_2 and RO_2, which greatly impacts the formation of HOM (Roldin et al., 2019; Nie et al., 2023) and changes the volatility distribution of the OVOCs and the resulting size dependent organic aerosol growth rates (Yan et al., 2020). To realistically represent how the SecA formation in the atmosphere influences the aerosol particle number size distributions (including UFPs) and the particle chemical composition we need atmospheric models that include temperature dependent atmospheric NPF parameterizations and chemistry mechanisms (including peroxy radical autoxidation mechanisms). These models need to dynamically update the volatility distribution of the condensable organic vapor according to the simulated ambient conditions (NO_x, RO_2, oxidant concentrations, temperature etc).

In PAREMPI we have, to our knowledge, for the very first time implemented a NPF formation rate lookup table approach that incorporates detailed NPF rate predictions through molecular modelling of sulfuric acid–ammonia nucleation into a global ESM (EC-Earthv3). This novel approach realistically represents how the temperature, atmospheric ion production rate, and molecular cluster scavenging sinks influences the NPF (Svenhag et al., 2024). This method is also used in the PAREMPI ePMI module. For the European scale air quality model simulations used for the Health impact assessment (HIA) in PAREMPI (see Sect. 5.2.1), we used the Lagrangian chemistry transport model ADCHEM (Roldin et al., 2019) (see PAREMPI **D3.3** report and result Sect. 4.2). This unique model explicitly represents the temperature dependent and ion mediated NPF involving NH_3, dimethylamine (DMA), H_2SO_4, methane sulfonic acid (MSA) and iodic and iodous acid (HIO_3, HIO_2) using the novel ClusterIn module (Olenius and Roldin, 2022; Wollesen de Jonge et al., 2024). The gas phase chemistry and secondary aerosol formation mechanisms, further developed in PAREMPI, are based on the master chemical mechanism (MCM) (Jenkin et al., 1997; Saunder et al., 2003; Bloss et al., 2005) which describes the tropospheric degradation of VOC. However, MCM's chemistry does not yet include autoxidation chemistry that result in the formation of HOM. Hence, ADCHEM also includes the unique Peroxy Radical Autoxidation Mechanism (PRAM) (Roldin et al., 2019; Pichelstorfer et al., 2024) including a complete, previously missing, oxidation mechanism for the monoterpene Δ^3-carene (Luo et al., 2024) and **new mechanisms for the formation of HOM from toluene (Iyer et al., 2023) and xylenes developed in PAREMPI.** However, despite of the complexity of the atmospheric chemistry mechanism (6425 species, 17970 reactions), it should be stated that even for the included most extensively studied monoterpenes and aromatic SOA precursors the detailed oxidation pathways and the resulting oxidation products volatility distributions are still highly uncertain especially at high NO_x-conditions and temperatures substantially below 20 °C.

1.5 Air quality, health and climate impacts of SecA from transport emissions

Air pollution and climate change are two of the most urgent societal challenges to humanity and the biodiversity on Earth and aerosol particles play a vital but highly uncertain role for both (.e.g. Szopa et al., 2021; Shrivastava et al., 2017; Landrigan et al., 2018).

From chemical and nano-physical point of view, $PM_{2.5}$ or UFP is not one pollutant, but a diverse group of aerosol particles, of different origin, composition, sizes and resulting impact on the air quality and climate on Earth. Depending on how the carbon atoms are ordered in carbonaceous aerosol particles, they may either primarily scatter or absorb sunlight (cooling or warming the climate). Although some secondary organic aerosols may absorb sunlight, especially UV light, and hence may sometimes be classified as brown carbon, secondary aerosols (both SOA and SIA) are primarily light scattering and hence have a direct net cooling effect on the climate on Earth (Shrivastava et al., 2017). Thus, carbonaceous aerosols are divided into mainly light scattering organic carbon aerosols (OA) and light absorbing black carbon aerosols (BC), where brown carbon (BrC) is an absorbing subfraction of OA. Similarly, the health impact and toxicity of carbonaceous PM depend on order of the carbon in the particles, ranging from highly reactive oxygen species (ROS) in secondary organic aerosols (SOA) to persistent solid BC particles that may stay and cause oxidative stress in the lungs and beyond for months. SecA, especially SIA, is also water-soluble by nature and hence both when SecA contributes to the formation of new aerosol particles via nucleation and growth and when SecA condenses onto hydrophobic primary aerosol particles such as diesel soot (Eriksson et al., 2017), it contributes to increasing the number, size and hygroscopicity of atmospheric aerosol particles, which is essential both for aerosol-cloud-climate-interactions and the particle deposition patterns in the respiratory system (Löndahl et al., 2007).

Aerosols profoundly impact climate, directly via aerosol-radiation interactions and indirectly via aerosol-cloud interactions (Szopa et al., 2021; Shrivastava et al., 2017). Aerosols affect cloud properties, lifetime and precipitation both by serving as cloud condensation nuclei (CCN) and as ice nucleating particles (INP). Most atmospheric aerosol particles with a diameter greater than 50-100 nm in diameter may serve as CCN at the right meteorological conditions (high enough water vapour supersaturation).

To properly assess the climate and health effects caused by aerosols, we need detailed knowledge of their chemical and physical properties and concentrations in space and time over the years. During the past 20-30 years, aerosol measurement techniques have advanced tremendously, and the pan-European research infrastructure network ACTRIS stations (>50 stations) offer year-round open access datasets for use by climate and air pollution scientists. However, they still provide limited spatial information about both the surface air pollution levels that the European population is exposed to and the concentrations in the vertical direction, which is critical for aerosol-cloud-climate interactions. Furthermore, although submicron particle number size distributions (PNSD) can be measured with mobility particle size spectrometers down to 2-3 nm in diameter, we have no complete measurement techniques to analyse the chemical composition of these aerosols. This is problematic since both the particle size and chemical composition, are essential for their toxicity, hygroscopicity, phase state (viscosity), light scattering and light absorbing properties, which in turn govern e.g. the direct and indirect aerosol RF, the hygroscopic growth and deposition patterns in the respiratory tract (Löndahl et al., 2007; Rissler et al., 2012), and health effects of the deposited particles (Landrigan et al., 2018).

Thus, there is a need for complementary theoretical methods that can be used to fill gaps in the spatial and temporal information about the aerosol concentrations and properties over the globe and which also can provide information about the chemical and physical properties of aerosols, over the complete climate and health relevant aerosol particle size spectrum from ~1 nm to >10 µm in diameter. Since the aerosol particle size distribution is vital both for the direct and indirect aerosol effects, the global climate and Earth System Modelling (ESM) community already work with models that can simulate the aerosol particle number size distribution evolution (aerosol dynamics) in the atmosphere (e.g. Svenhag et al., 2024, 2025). But this is challenging since aerosol dynamics models are computationally expensive and the dynamics changes on timescales of a few seconds. Thus, it is always a trade-off between how detailed aerosol dynamics can be represented and the spatial

resolution of the models. In the global chemistry transport model TM5, which is implemented in the European community Earth System Model version 3 (EC-Earth3; Döscher et al., 2022), the aerosol dynamics is represented by the M7-modal scheme (Vignati et al., 2004) with, 4 soluble and 3 insoluble modes, and the SOA formation is represented with two species, one SVOC and one ELVOC (Bergman et al., 2022).

From the air quality modelling community less attention has been paid to aerosol size distributions and aerosol dynamics. One reason is likely that the air quality standard in EU and worldwide are based on the mass concentrations of particulate matter less than 2.5 µm in diameter ($PM_{2.5}$) and less than 10 µm in diameter (PM_{10}). Hence, air quality models generally do not consider aerosol dynamics. However, with the emerging concern about the health effects caused by ultrafine aerosol particles (UFP), i.e. particles less than or equal to 100 nm in diameter, some air quality modelling groups have started to model UFPs (e.g. Frohn et al., 2021; Kohl et al., 2023). The new EU air quality directive (DIRECTIVE (EU) 2024/2881) which will require BC and UFP monitoring at urban supersites, will likely also increase both the measurements and the modelling studies of UFP in the future.

Transport emissions, especially from road traffic, are important sources of NO_x, VOCs and to less extent NH_3 and SO_2, which contribute to the SIA, SOA and the tropospheric ozone levels in Europe (EMEP, 2023). Organic aerosol (OA) is the dominant source non-refractory PM_1 (NR-PM_1) in most regions in Europe, followed by sulfate and nitrate. However, during episodes with elevated NR-PM_1 levels exceeding 40 µg m^{-3}, nitrate often dominates the NR-PM_1 (Bressi et al., 2021). According to the atmospheric model simulations performed with the chemistry transport model ADCHEM in PAREMPI (PAREMPI **D3.3** report), the road traffic in Europe is primarily contribution to SecA in the form of nitrate PM (see Sect. 4.2) and to lesser extent SOA. However, most likely that the model still underestimates the SOA formation from many anthropogenic VOCs since we presently only consider peroxy radical autoxidation leading to HOM SOA for a handful of aromatic VOCs, and the chemistry, especially at high NO_x conditions, is still highly uncertain (see Sect. 1.3).

1.6 Identification of gaps in knowledge

Despite growing awareness of the role of traffic in SecA formation, significant gaps remain in our understanding, limiting the accuracy of emission inventories and the effectiveness of mitigation strategies. These gaps span across several critical areas. Firstly, combustion produces large amounts of different precursor compounds, which remain poorly characterized and quantified, inhibiting creating a detailed understanding of the SecA formation process. One of the most fundamental uncertainties lies in the identification and modeling of organic precursors. The chemical complexity and variability of these compounds make it difficult to predict their transformation into secondary aerosols. Current models lack robustness, and the contribution of specific precursors remains poorly quantified.

PART II: Quantification and effects of aged PM from transport sources

2 Methods for quantifying PM_{aged}

2.1 Features of measurement system for PM_{aged} and precursor gases

2.1.1 Key components required for quantifying PM_{aged} emissions

The PAREMPI project measurement campaigns were designed to investigate the formation of secondary aerosols in real-driving and field conditions. This approach highlights the need for a robust measurement system capable of accurately quantifying aged PM emissions and gaseous precursors that contribute to secondary aerosol formation. It also acknowledges the need for methods to estimate harmfulness of $PM_{2.5}$ in portable setups. The ambitious aim in the PAREMPI was to design a robust measurement system, and to use a comprehensive state-of-the-art aging system and the novel PAREMPI robust measurement system in parallel, which increased complexity of the measurements further.

Regardless of the complexity of the measurement system, multiple phases from the raw exhaust to the PM_{aged} results are involved. Table 1 provides a schematic overview of the measurement pathway and its key components, including sampling, dilution, precursor gases, oxidation flow reactor (OFR), precursor gases, PM_{aged}, and toxicity assessment. The detailed measurement configuration is described in Section 2.1.4, while the proposed robust measurement system is outlined in the PAREMPI **D2.3** report.

Table 1. Key elements of robust measurement system to be considered.

Sampling	Gases	Dilution	OFR	Aged PM
•Sampling guidelines, i.e. need for heated raw exhaust transfer lines.	•Precursor gases. •Gases needed to define e.g. OFR settings.	•Dilution ratio. •Requirements for dilution system.	•Requirements for the OFR. •Settings for the OFR.	•Methods to determine PM. •Emission factors •Toxicity.

Sampling, dilution and precursor gases

Accurate sampling of raw exhaust is essential for reliable and representative measurements. Exhaust needs to be transferred to the first diluter without cold spots to avoid condensation of water, sulfuric acid, or organic compounds. Heated transfer lines maintain exhaust unaltered. While standards guide sampling, applying them in real-world conditions remains challenging. Key recommendations include a suitable probe type and line setup, leak inspections, monitoring of temperatures of heated lines, personnel trained for installations and measurements and sufficient power management.

To prevent nozzle clogging, mild heating (~50 °C) of the first ejector stage may be applied; for high-sulfur marine engine exhaust, heating to approx. 200 °C is advised. Placing dilution close to the sampling point reduces energy demand. After dilution, temperatures can be lower. Short, straight sampling lines minimize particle losses.

Measuring SecA precursor gases (NO, NO_2, NH_3, SO_2 and VOCs, particularly aromatics) is needed, as well as NO_x and CO concentrations for the OFR settings, and CO_2 for PM_{aged} calculation.

2.1.2 OFR for simulation of PM_{aged} formation potential

2.1.2.1 General

Real-time measurement methods for assessing the secondary aerosol formation potential of exhaust emissions are typically conducted using oxidation flow reactors (OFRs) such as the Potential Aerosol Mass (PAM) or the Dekati oxidation flow reactor (DOFR). In the OFRs, irradiation with UV light is used to form high concentrations of oxidants (O_3, OH, HO_2) which ensure fast oxidation of compounds and initiation of SecA formation (Figure 3). The settings of UV light (wavelengths, intensity) are selected to represent atmospheric aging of emissions from minutes to weeks as the sample flows through chamber, and consequently, these settings affect substantially the quantity of SecA formed in the OFR. To ensure that oxidation occurs within minutes or even seconds, the reactor contains 100-1000 times the amount of OH radicals compared to the lower atmosphere (Kang et al. 2007). The progress of the oxidation reactions in the OFR is described by the photochemical age, which corresponds to the time spent in the oxidant concentration of the lower atmosphere.

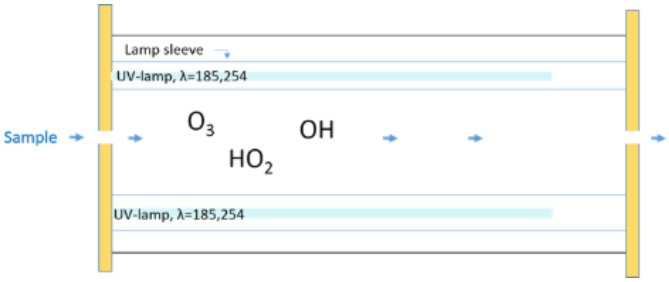


Figure 3. A schematic presentation of a PAM oxidation flow reactor (OFR).

Several types of OFRs have been adapted into use by the research community. The size of chambers varies from very small, 0.15-liter chamber with residence time of seconds (Keller and Burtscher, 2012) to very large chambers with volume up to 155 liters (Ihalainen et al., 2019). In general, the residence time depends on the flow through chamber and the volume of the chamber. For atmospheric studies or laboratory studies with stable measurement points, the chambers like the Potential Aerosol Mass (PAM, Aerodyne Research Inc.) chamber with larger volume and longer residence time are shown to give representative results (Lambe et al., 2011). The PAM chamber, made of aluminium, has a volume of 13.3L and two Hg-lamps to produce oxidants (OH, O_3, HO_2). The amount of oxidants is high to mimic atmospheric aging in minutes the sample goes through the chamber. Chambers with shorter residence times are required for emission studies using transient emission sources. For this end, TSAR, Go:PAM, and MSC have been developed (Simonen et al., 2017, Watne et al 2018, Keller et al., 2012). Zhang et al., (2024) provide a summary of the most commonly used OFRs. Review by Ma et al., 2025 overviews existing OFRs and compares their structural differences and compares results between studies.

2.1.2.2 Aging period

Aging period at the OFR must be selected based on photochemical considerations and a comprehensive assessment of previous studies to support the chosen timeframe in which the maximum potential aerosol mass is achieved. Specifically, Xu et al. (2022) reported that secondary organic aerosol (SOA) formation at urban roadsides occurs over 1–2 days of photochemical aging, which is shorter than the approx. 3 days observed in forests dominated by biogenic VOCs (Slowik et al., 2012) and within the 0.8–6 day range found in urban Los Angeles (Ortega et al., 2016). According to Monod et al. (2001) the atmospheric lifetime of most significant anthropogenic aromatic compounds from engine emissions ranges between 11.8 hours and 2.9 days, with the exception of benzene, which persists up to 9.4 days (assuming an OH concentration of 10^6 rad cm^{-3}), supporting the SOA formation period reported by Slowik et al. (2012). Therefore, the majority of the most relevant VOCs from engine emissions would be nearly fully oxidized within three days. Additionally, oxidation products can partition into the particle phase quickly, as shown in a study by Monge et al. (2012) that has reported typical atmospheric particle growth rates between 1 and 20 nm h^{-1} using data from over 100 field studies, with consistent daytime values. After most VOCs have undergone oxidation and equilibrium between gas and particle phases has been reached, further SOA formation from precursor VOCs emitted by a specific engine becomes minimal, while particle losses due to decomposition and deposition become more relevant. As a result, a 2–3 day maximum aging period is a reasonable upper limit for engine emission experiments using OFR, beyond which particle sizes may also become less relevant from a toxicological perspective due to further growth e.g. through condensation or coagulation.

In the PAREMPI, **a target maximum aging of 2–3 days** at the OFR was selected.

2.1.2.3 Comparison of PAM and portable OFR in PAREMPI

As mentioned, OFRs are essential for measuring PM_{aged} (Sections 2.1.1-2.1.2) with portable models now available and easier to operate via advanced control software. Despite the simplified operation, a protocol for setting measurement parameters, including dilution, aging period, and verification, is needed for consistent results. In the PAREMPI exhaust emission measurement campaigns, two different OFRs, PAM and DOFR, were used to measure the PM_{aged} formation potential.

Aging in OFRs depends on humidity, ozone, temperature, UV intensity, and residence time, which is verified using tracer gases (e.g., CO, SO_2, d_9-butanol). These tracers help monitor OH depletion, particularly under high NO_x or VOC conditions. Dilution before OFR is crucial to avoid oxidant depletion, but excessive dilution can hinder detection. Post-OFR dilution supports flow stability and chamber cleaning, which are faster in smaller reactors.

Optimizing the OFR settings is key for simulating atmospheric SecA formation. Target NO_x concentrations should be in the ppm range to avoid altering oxidation chemistry (Peng et al., 2015). Dilution needs to vary by exhaust source. Tracer gases add complexity but are currently the best option; modeling based on gas concentrations and OFR settings may offer alternatives. Background cleaning and zero-air measurements are also essential.

In PAREMPI, the Dekati Oxidation Flow Reactor (DOFR) was used. Compared to PAM, the DOFR offers a shorter residence time, higher oxidant levels, and better time resolution, making it suitable for transient exhaust conditions. It includes built-in flow control, humidity adjustment, ozone generation, and sensors. DOFR's laminar flow minimizes particle losses. While user-friendly, finding optimal settings remains challenging and may introduce variability.

In the PAREMPI, DOFR and PAM were compared in LD car tests for 7 vehicles at three different temperatures. An example of temporal variation of PM_{aged} from CNG car emission measurements

with the DOFR and PAM chambers is shown in Figure 4. Results showed DOFR's higher time resolution captured emission dynamics better during driving events. Lower PM_{aged} concentrations in DOFR may result from reduced wall losses and background levels compared to PAM. Differences in particle resuspension and residence time likely explain observed variations.

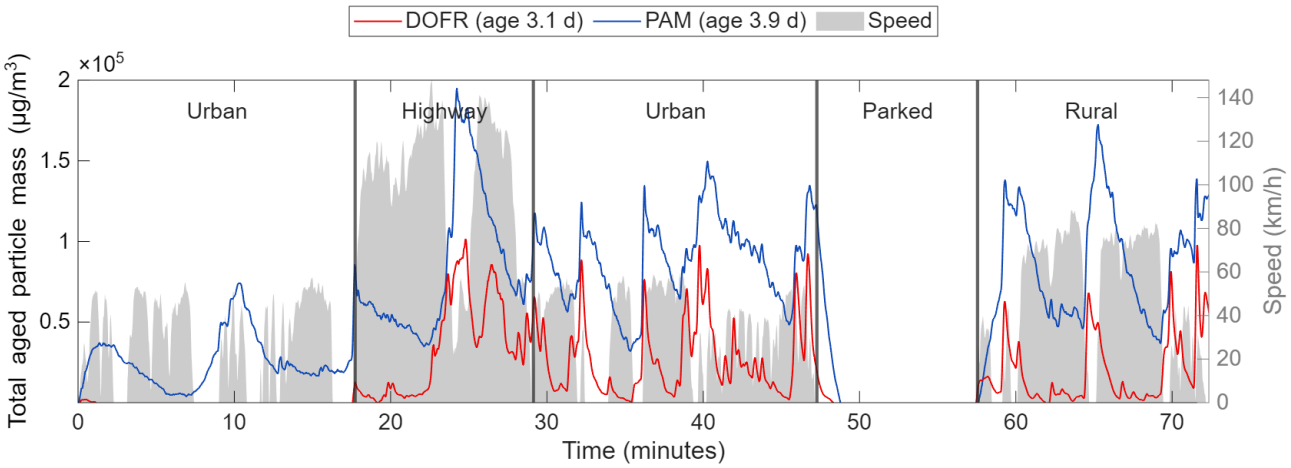


Figure 4. An example time series of total PM_{aged} particle mass measured with parallelly installed DOFR (red) and PAM (blue) for the CNG car. Dilution corrected to raw exhaust. Legend shows atmospheric equivalent age in the chambers. PM_{Aged} contains primary and secondary particles.

2.1.3 Measuring PM_{aged} and calculation of emission factors

After the OFR, either chemical composition with AMS and aethalometer or size distribution of particles with ELPI was measured to monitor the PM_{aged} concentration and composition in realtime in PAREMPI experiments. The online PM monitoring gives important information about the temporal variation of PM_{aged} formation during different driving conditions such as cold start, acceleration or deceleration.

For robust setup described in Section 6.4, the determination of PM_{aged} mass emissions is suggested to be based on the size-distributed particle (PSD) number concentration, converted to mass assuming spherical particles with unit density. Instruments like an ELPI and SMPS are commonly used. The ELPI measures aerodynamic diameters (7 nm-10 µm) and provides real-time number and mass distributions, whereas the SMPS measures electrical mobility diameters (2 nm-1 µm). The ELPI is preferred for OFR studies due to its broader range and temporal resolution. The AMS is also widely used for real-time chemical composition (e.g., NO_3^-, SO_4^{2-}, NH_4^+, organics), though it excludes black carbon (BC), which requires separate analysis.

For the calculation of PM_{aged} emission factors by using exhaust flow or CO_2 data and data measured after the OFR, the rapid changes measured from the tailpipe are smoothed in the OFR due its relatively long residence time and mixing distortion needs to be synchronized. Simonen et al. (2024) proposed a convolution method to model OFR response, while PAREMPI used a moving average based on residence time, with both methods showing good correlated (Figure 5). This procedure is distinct from the time alignment of data from different instruments.

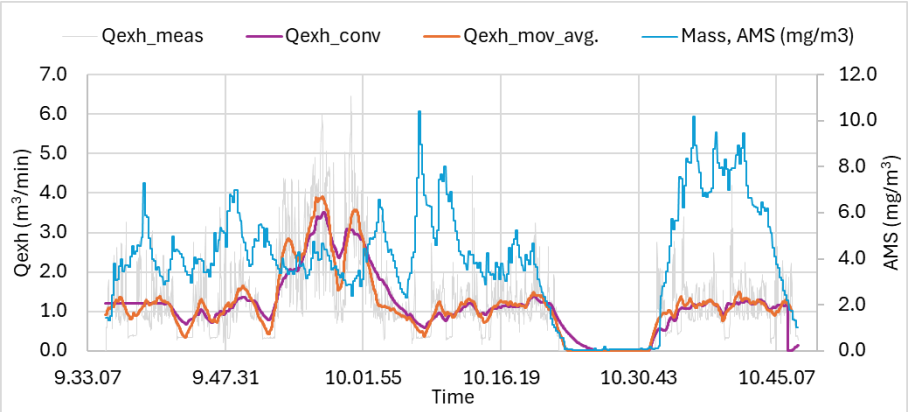


Figure 5. Exhaust flow rate measured upstream the OFR (Qexh_meas, grey) and calculated by convolution (Qexh_conv, purple) and moving average (Qexh_mov_avg, orange) methods to represent exhaust flow downstream the OFR. Sub-micron particle mass measured by AMS (Mass, AMS, blue) excludes black carbon.

To define PM_{aged} emission factors consistently across transport sectors, a unified metric for the results is essential. Currently, emission factors are expressed in varying units depending on the sector. PAREMPI recommends using mass per megajoule (mg/MJ), which simplifies the setup by eliminating the need for exhaust or fuel flow measurements and enables harmonization across transport sectors. Energy unit refers to energy consumed, either fuel or electricity.

Emission factors per MJ are calculated by dividing exhaust species concentrations by CO_2e (including CO_2, CO and organics), then converting CO_2e to fuel and energy consumed using known fuel carbon and energy content. This approach enhances robustness, especially in aviation and marine sectors, where exhaust flow measurements are challenging. However, this method applies only to carbon-based fuels; future carbon-free fuels will require alternative markers.

Although not included in the robust measurement system described in Section 6.4, measuring the PM_{fresh} emissions is recommended besides the PM_{aged} emissions, preferably the regulated PM used in inventories. This would enable evaluation of ratio between PM_{aged} and PM_{fresh}(reg), and hence, SecA formation potential. This ratio is illustrative of the differences between applications and fuels, since these ratios may be low (e.g. 1-5) or high (>10) depending on case (Suarez-Bertoa et al., 2015, Timonen et al., 2017). Additionally, measurements of the other exhaust characteristics, such as semi-volatiles, polycyclic aromatic hydrocarbons (PAHs), anions, BC, organics and metals, would enhance understanding.

PM_{fresh} measurements are typically done from diluted exhaust, by collecting PM on filter, which is subsequently weighed to determine mass. However, other instruments capable of measuring PM or PN concentrations in realtime can provide more detailed information about PM, for example the temporal variation during different driving conditions such as cold start or acceleration.

2.1.4 PAREMPI measurement systems for PM_{aged} and precursor gases

The campaigns were performed by international collaboration involving VTT Technical Research Center of Finland, Tampere University, Finnish Meteorological Institute, BOSMAL Automotive Research and Development Institute, Institute of Experimental Medicine, ONERA French Aerospace Lab and Lund University. Comprehensive emission characterizations were conducted in the measurement campaigns of the PAREMPI project. In all four exhaust measurement campaigns versatile research-grade instrumentation covering both gaseous and particulate pollutants as well as OFRs for PM_{aged} formation potential simulation were used (Figure 6). Toxicity experiments were linked with emission characterizations, described in deliverable report **D2.3 and D4.2** of the project.

One of the main research topics of the experiments was to evaluate the potential of exhaust from internal combustion engines to form secondary aerosols in the atmosphere. This was investigated by utilizing oxidation flow reactors for the exhaust sample flows, incorporated into the measurement setups with appropriate exhaust sampling and dilution systems and instruments that provided measurement data for the concentrations of SecA precursor gases and aerosol particles. Based on the experience and knowledge collected in the project, the SecA forming potential of exhaust can be measured repeatably and so that e.g. the effects of vehicle driving conditions and vehicle technologies on SecA formation potential can be quantified. The SecA measurement technology point of view of the project is included in the separate report, Deliverable **D2.3**.

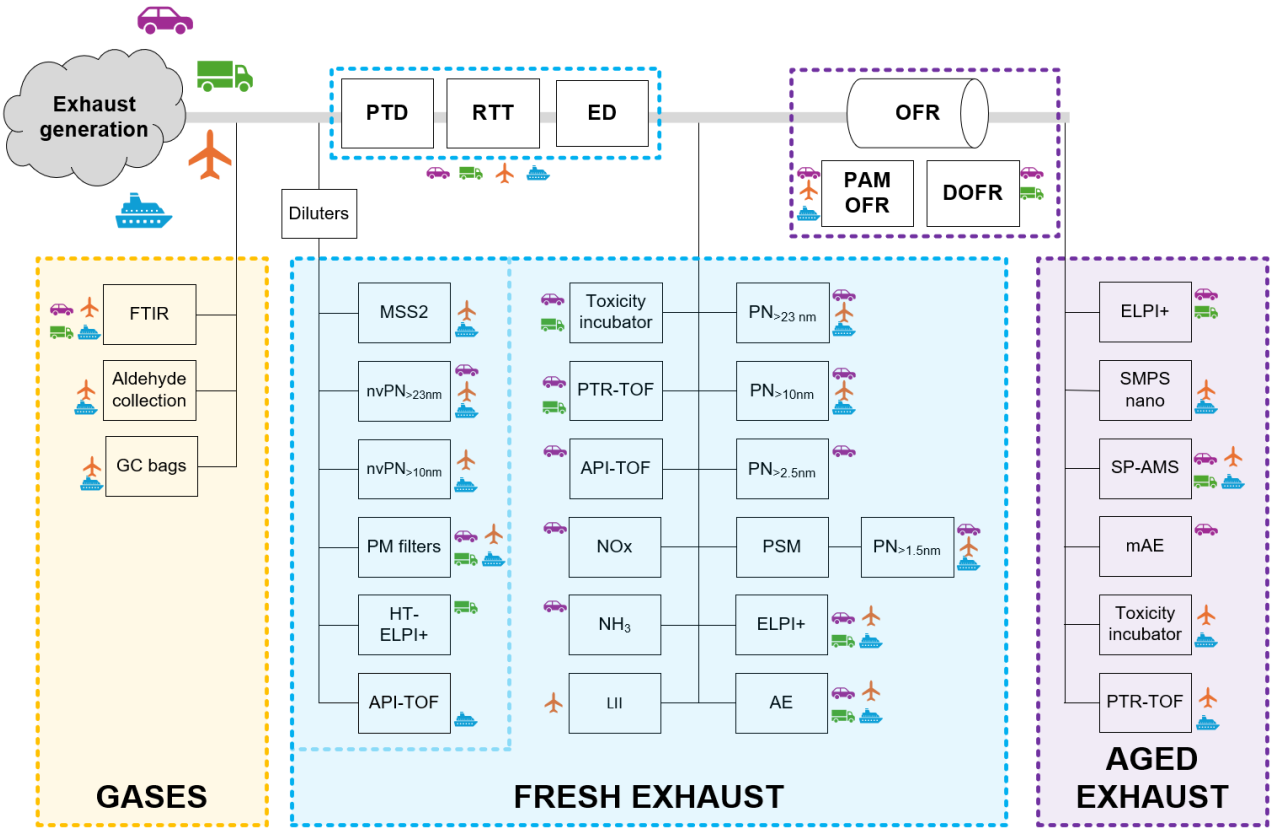


Figure 6. Generalized schematic of PAREMPI set-ups. With each instrument/metric/part of the schematic, symbols (car, truck, airplane, ship) indicate the experiment where it was used. Abbreviations: AE = Aethalometer, API-TOF = Atmospheric Pressure Interface Time of Flight, DOFR = Dekati Oxidation Flow Reactor, ED = Ejector Diluter, ELPI = Electrical Low Pressure Impactor, FTIR = Fourier Transfer Infrared, GC = Gas Chromatograph, HT = High Temperature, LII = Laser Induced Incandescence, mAE = MicroAethalometer, MSS = Micro Soot Sensor, NH_3 = Ammonia, NOx = Nitrogen Oxides, nvPN = Non-Volatile Particle Number, OFR = Oxidation Flow Reactor, PAM = Potential Aerosol Mass, PM = Particle Mass, PN = Particle Number, PSM = Particle Size Magnifier, PTD = Porous Tube Diluter, PTR-TOF = Proton Transfer Reaction Time of Flight, RTT = Residence Time Tube, SMPS = Scanning Mobility Particle Sizer, SP-AMS = Soot Particle Aerosol Mass Spectrometer.

The robust measurement system for quantifying aged PM emissions and gaseous precursors forming secondary aerosols suggested by PAREMPI is presented in Section 6.3 and in **PAREMPI D2.3 report.**

2.2 Description of PAREMPI measurement campaigns

Based on the initial literature database and unpublished data from author organizations, gaps in the knowledge were identified to conduct complementary measurements of PAREMPI (confidential **D2.1** report). For each sector (light-duty, heavy-duty, marine, aviation) the current fleet and fuels in use were briefly analyzed. Common measurement methodology appliable to all sectors and all campaigns were designed, and sector-specific measurement approaches were also discussed. The results from experimental campaigns aimed to supplement and extend existing knowledge.

Five intensive measurement campaigns were conducted in the PAREMPI project (Figures 6 and 12). Two of the campaigns focused on exhaust from light-duty passenger cars and heavy-duty trucks, two utilized laboratory setups to mimic emission phenomena related to marine engine exhaust and aircraft engine exhaust, and one campaign investigated brake dust emissions. The experimental campaigns aimed to generate valuable insights into fresh and aged particulate emissions as well as gaseous precursor emissions from the transportation sectors, addressing significant knowledge gaps in these areas. A detailed description of measurements can be found from the PAREMPI deliverable **D2.2.** Toxicity experiments were linked with emission characterizations, described in deliverable reports **D2.3** and **D4.2** of the project. Measurement setup for experiments to evaluate the potential of exhaust from internal combustion engines to form secondary aerosols in the atmosphere are described in Section 2.1.4.

Table 3 includes a summary of vehicles, technologies and fuels used in the PAREMPI exhaust measurement campaigns. Shortly, in the **LD campaign** to reflect the EU light-duty vehicle market, a diverse selection of cars was tested, including plug-in hybrids, non-hybrids, gasoline, diesel, Euro 6, and older models (Vehicles: PHEV-G-EU6d (V1), PHEV-D-EU6d (V2),G-EU6d (V3), D-EU6d (V4), G-EU6b (V5), D-EU4 (V6), CNG-G-EU6d (V7).) All vehicles had direct injection engines and relatively low mileage, representing popular global or local brands. Fuels used met EN228 (gasoline) and EN590 (diesel) standards, with winter-grade diesel used at –9°C. A bi-fuel CNG vehicle primarily ran on Finnish pipeline CNG but used EN228 gasoline during start-up and heavy loads, as per manufacturer specifications. The RDEsim test cycle was developed to mimic real-driving emissions (RDE) under extended and repeatable conditions in a laboratory setting. Based on the Euro 7 proposal COM(2022) 586, the cycle was designed to be applicable across various ambient temperatures and vehicle types. A reference on-road RDE test, known as the "golden PEMS RDE test," was conducted first and this cycle was then replicated on a chassis dynamometer for all tested light-duty vehicles. Emission measurements were carried out at BOSMAL's Emission Laboratory No. 2 using a 4WD AVL Zoellner 48" dynamometer and a WEISS WK 643 climatic chamber, with tests performed at –9°C, +23°C, and +35°C. The Aged PM results of the campaign have been described in the publication by Simon et al., (2025). Several other publications about the results are currently being prepared. For the **HD campaign,** two trucks were selected for the winter-time heavy-duty truck emission measurement campaign of the project. The newer truck conformed to Euro VI E emission standard and had the odometer reading of 65465 km, and the older truck conformed to Euro VI A standard and had odometer reading of 1207725 km. Two types of fuels were utilized in the heavy-duty measurements: fossil diesel fuel (F), which complies with the EN590 standard, and renewable diesel (R), which meets the EN 15940 standard. Notably, the fossil diesel fuel did not contain any fatty acid methyl esters (FAME). The instrumentation was placed on the seacontainer, placed on the trailer behind the truck. Electricity and pressurized air were produced with instruments placed on the trailer. On-road experiments with two trucks covered various distances (80-500km) and routes in middle and northern Finland in winter conditions. In the experimental campaign focusing on **marine exhaust**, the exhaust was generated in the laboratory facilities to mimic the characteristics of marine engine exhaust. Simulation of different marine exhaust compositions were conducted by using a diesel generator from which exhaust aftertreatment system was removed prior to the measurements

and marine fuels. Fuels included DMB, DMB with doping agents or injection of chemicals to produce initial exhaust obtained with fuels with high sulfur, ash, or ammonia content, and other fuels covered EN590 and HVO. Ammonia was fed to the exhaust after the diesel generator, using in-house-developed system based on water-ammonia mixture and evaporation method to achieve three ammonia concentrations in the exhaust. A permeation oven (FlexStream Base, Kin-Tek Laboratories, Inc., USA) was used to generate hydrocarbon gas mixtures—benzene, p-xylene, naphthalene, dodecane, and toluene—relevant to real-world exhaust emissions and SOA formation. The mixtures were produced in a 1 L/min nitrogen flow and combined with actual exhaust. Commercial permeation tubes (calibrated at 80 °C) were used for all compounds except toluene, which was delivered via a custom tube calibrated at 100 °C and tested separately. Once stable concentrations were confirmed via Vocus-PTR-MS, the mixtures were introduced into an oxidation flow reactor under both dark and illuminated conditions to study SOA formation.

The CAST generator was used in **aviation emission studies** due to its low fuel consumption (a few ml/h) compared to full-sized aircraft engines and thus better possibilities to conduct experiments with various fuels and fuel mixtures. In the aviation fuel experiment, seven different fuels were tested, representing commercial fossil jet fuels and sustainable aviation fuels (SAF) containing synthetic paraffinic kerosene (SPK) component SBC (synthetic blending component) (Table 2). Three fuels were fossil Jet A-1 fuels (J1, J2, J6), two fuels contained a SPK SAF component (J3, J5) and one fuel was 100% SAF SPK jet fuel (J7). Fuels of the matrix had varying sulfur and aromatic contents.

Table 2. Summary of vehicles, technologies and fuels used in the measurement campaigns.

Exp.	Code	Technology	Fuel Type
LD	PHEV-G (V1)	PHEV, Gasoline, Euro 6d	EN228 Gasoline
LD	PHEV-D (V2)	PHEV, Diesel, Euro 6d	EN590 Diesel (summer/winter grade)
LD	EU6d-G (V3)	Gasoline, Euro 6d	EN228 Gasoline
LD	EU6d-D (V4)	Diesel, Euro 6d	EN590 Diesel (summer/winter grade)
LD	EU6b-G (V5)	Gasoline, Euro 6b	EN228 Gasoline
LD	EU4-D (V6)	Diesel, Euro 4	EN590 Diesel (summer/winter grade)
LD	EU6d-CNG (V7)	Bi-fuel CNG-Gasoline, Euro 6d	Finnish pipeline CNG (primary), EN228 Gasoline (secondary)
HD	EU VIE (Truck 1)	Euro VI E	EN590 Fossil Diesel (no FAME), EN15940 Renewable Diesel
HD	EU VIA (Truck 2)	Euro VI A	EN590 Fossil Diesel (no FAME), EN15940 Renewable Diesel
Marine	DMB, DMB+S, DMB+ash, DMB+NH_3, EN590, HVO	Diesel Generator (no aftertreatment)	DMB, DMB high-sulfur, DMB ash-forming component, NH_3 injection in exhaust, EN590 Diesel, HVO
Aviation	J1-J7	Cast combustion system and oil generator	Jet A-1 (typical market fuel), Jet A-1 (higher S and naphthalene, lower aromatics), two HEFA-SPK blends, F-35, FT-SPK (Fischer-Tropsch Synthetic Paraffinic Kerosene)
Other		Permeation oven experiments	Benzene, p-xylene, naphthalene, dodecane, and toluene

2.3 Identification of gaps in knowledge on measurement systems

Preventing secondary aerosol formation from transport emissions requires addressing several key knowledge and technology gaps. One major challenge is the **limited availability of measurement**

devices that are suitable for real-world and on-road conditions. Without portable and reliable tools to monitor SecA precursors and formation in the field, it is difficult to evaluate the effectiveness of mitigation strategies. The robust measurement system proposed by PAREMPI represents an advancement in expanding the capabilities for measuring aged particulate emissions from transport sources.

Regardless of complexity of the measurement setup, critical issue in exhaust studies is the **lack of standardized protocols** for sampling, dilution, oxidation flow reactors (OFRs) and subsequent PM measurement principles and calculation methods. The OFR reactors are essential for simulating atmospheric aging processes, but without consistent settings (e.g., flow rate, dilution, oxidation state), lead to variability across studies. Establishing a **Standard Operating Procedure (SOP)** for OFR use, including agreed operating conditions, validation and calibration procedures, and cleaning instructions, would enhance comparability. Although these protocols fall outside the scope of PAREMPI, they are necessary actions that should be addressed to harmonize measurement conditions across laboratories.

The PM_{aged} results obtained are rather semi-quantitative regardless of the complexity or robustness of the measurement system due to the sensitivity of the many uncertainties in the measurements. In addition, a variety of different OFRs with different conditions are used when simulating PM_{aged} formation, complicating the comparison between studies. Furthermore, different calibration schemes for OFR are used and the results are given in a variety of different units. However, despite the differences the results indicate that when SecA precursors are substantially present in exhaust, the PM_{aged} typically increases to many-fold levels when compared to PM_{fresh} emissions.

Development of remote measurement systems, such as drones, sniffers, roadside measurement systems or even satellite measurement would enable realtime monitoring of traffic streams and their emissions, including SecA precursors, such as NO_2 and SO_2, and detection of polluters and non-compliant systems remotely without conducting tedious and time-consuming RDE, on-board and field measurements. However, we note that **measuring PM_{aged} remotely in the realworld conditions is likely impossible, due to the dilution of exhaust and mixing of emissions with atmospheric pollutants during the transport from the source to OFR. The PM_{aged} measured would not represent the original source**.

3 Results from PAREMPI campaigns

3.1 Results from emission measurement campaigns

3.1.1 Overview of results

Results of exhaust experiments of the PAREMPI project show the significance of PM_{aged} formation potential in respect of the total atmospheric impact of exhaust from different transport modes, which may have implications also to the total air quality and health impacts of traffic exhaust. Furthermore, the PM_{aged} formation from exhaust emissions was found to be dependent on many technology-related parameters, such as fuel and exhaust aftertreatment. In the experiments with light-duty and heavy-duty vehicles, especially cold starts of engines were identified as important in respect of exhaust emissions, including their contribution to the SecA formation potential of exhaust. SecA was linked especially with gaseous organic compounds that typically originate from fuel and lubricant oil, but also with inorganic gaseous pollutants SO_2, NO_x and NH_3. Thus, the total SecA impact of emissions to air quality cannot be reduced only by focusing on one technology. In addition, it is

important to notice that e.g. the most modern vehicles emit relatively low amounts of regulated pollutants, which may increase the relative role of currently unregulated emissions – particularly those with the potential to form SecA in the atmosphere. Results from light-duty vehicle experiments indicate also that the relative role of the smallest particles, i.e., currently unregulated sub-10 nm particles found from fresh exhaust, can be higher with modern vehicles than with the older ones. Additionally, even for the most modern cars and trucks, PAH species were found in exhaust. On the other hand, also the high efficiency of exhaust emission reduction techniques was proved in the experiments, seen especially as low black carbon emissions from vehicles equipped with exhaust filtration devices. Overall, in addition to SecA formation potential and nano-sized particles of exhaust, also the potential of PAH found in the exhaust of modern cars and trucks to pose harmful health effects emphasizes the need for further development of fuels, lubricating oils, engines and aftertreatment technologies to mitigate these emissions.

Here we present findings from PAREMPI measurements, database and selected supporting literature to enlighten possibilities to prevent SecA and PM_{aged} emissions from transport sectors by technological solutions. In the end of this section, evaluation and conclusions are summarized in Table 4.

3.1.2 SecA precursors and PM_{aged} emissions

The results of measurement campaigns in PAREMPI are summarized in Figure 8 for precursor gases and PM_{aged}, and in Figure 9 for composition of PM_{aged}, across vehicles, engines, fuels and exhaust aftertreatment systems, together with reference values. Emission factors of PM_{aged} are presented in harmonized unit (mg/MJ) to enable cross-sectoral comparison.

Cars

The PAREMPI results highlight how tightening emission standards have decreased exhaust emissions from cars. The results published by Simon et al. (2025), revealed a downward trend in both PM_{aged} and SecA precursor emissions as regulations have become more stringent. Among these precursors, NO_x emissions are directly regulated, while SO_2 is indirectly controlled through road fuel standards (EN228 and EN590). NMOG and NH_3 remain unregulated for passenger cars in Europe, highlighting regulatory gap with implications for SecA formation.

The PAREMPI study found that new Euro 6d gasoline and diesel cars emitted significantly lower PM_{aged} compared to older Euro 6b gasoline, Euro 4 diesel, and Euro 6dT CNG/gasoline. Interestingly, while emission standards appeared to indirectly reduce PM_{aged}, Euro 6dT CNG car emitted high PM_{aged} emissions attributed to the gasoline backup system, which contributed to emissions in certain driving conditions.

SecA formation was observed in nearly all studied cars, with the exception of the Euro 4 diesel, which exhibited high PM_{fresh} and PM_{aged} emissions. Although new cars emitted lower overall emissions, cold-start conditions remained a challenge, also triggering SecA formation. PHEVs added another consideration: when transitioning from electric to combustion mode, they emitted elevated particle concentrations, especially under dynamic driving conditions.

PM_{fresh} emissions were notably low for modern cars with filtration systems. Diesel vehicles with DPFs emitted very low particle emissions, though regeneration events may occasionally lead to elevated emissions. Gasoline car lacking GPFs still emitted high particle number emissions, especially in cold conditions.

While TWCs are effective at reducing NO_x and NMOG emissions, they can emit NH_3 as a byproduct, enhancing SecA formation through promoting ammonium nitrate (NH_4NO_3) or ammonium sulfate

($(NH_4)_2SO_4$) formation, depending on exhaust composition. Gasoline cars may have a significant role for SecA formation in NH_3-limited urban environments (Simon et al. 2025), and future emission standards for cars would benefit from NH_3 limits.

Heavy-duty vehicles

In the PAREMPI study, two heavy-duty (HD) trucks were studied, an older Euro VI A model and a newer Euro VI E, both equipped with modern exhaust EAT systems. The results for the new Euro VI E truck demonstrated exceptionally low PM_{aged} emissions. Unfortunately, PM_{aged} measurements for the Euro VI A truck were not complete. Still, the data confirms an important role of EATS in reducing PM_{aged} emissions from HD engines.

As regards SecA precursor emissions, the Euro VI E truck again performed well. Both NO_x and NMOG emissions were below the upcoming Euro 7 limits, demonstrating the effectiveness of its emission control. In contrast, the older Euro VI A truck exhibited elevated NO_x emissions. Notably, NH_3 emissions from both trucks remained low, staying within the Euro VI limit of average 10 ppm. This suggests that SCR systems were functioning efficiently, possibly in combination with the ammonia slip catalyst.

Emission factors for trucks were low over the long driving distances of 200 km and 80 km due to the dilution effect masking the cold start emissions. For both trucks, cold start at winter temperatures induced high emissions before the EAT systems warmed up (Figure 7).

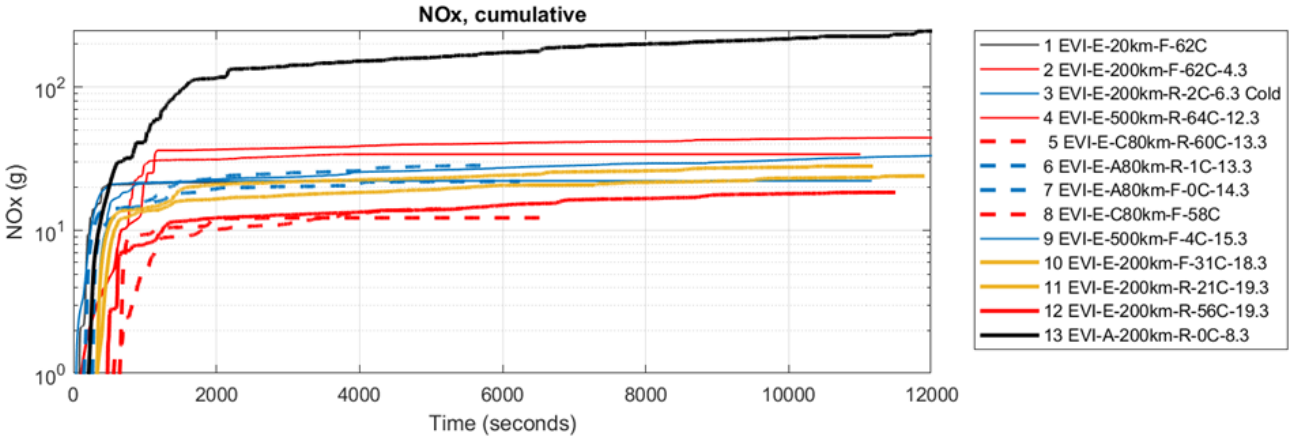


Figure 7. Cumulative NO_x emissions with two trucks, Euro VI A and Euro VI E.

Marine

The PAREMPI experiments using a diesel generator and marine fuels revealed a wide range of aged particulate matter, alongside fluctuating levels of precursor gases. These measurements with diesel engine were conducted without any EAT systems. Both fresh and aged PM emissions were substantial.

In PAREMPI experiments, NO_x emissions were substantial, but at slightly lower than the average reviewed for certain type of marine engines (Aakko-Saksa et al., 2023).

An interesting case was an intentional addition of NH_3 by injection in the exhaust at level of 60 ppm, shifting the composition of PM_{aged} remarkably. Nitrates became the dominant component, accompanied by smaller amounts of ammonium and organics, suggesting clear formation of SecA, particularly ammonium nitrate and organic nitrates. Overall, this NH_3 addition led to the most significant SecA formation in the marine engine simulation underscoring its role in driving secondary aerosol reactions. When NH_3 was not injected in exhaust, NH_3 levels remained near the detection limit.

Again, when marine fuel contained close to 0.5 wt-% sulfur, composition of PM_{aged} shifted to sulfates, pointing to sulfur-driven SecA formation. SO_2 emissions expectedly followed the sulfur content of the fuels.

When using distillate marine fuel, DMB, heavy fractions originating fuel formed PM through condensation. NMOG emissions were high when using marine fuels, higher than those from passenger cars and heavy-duty vehicles, aligned with supporting literature.

Aviation

In the PAREMPI simulations using a CAST burner and aviation fuels, both fresh and PM_{aged} emissions were found to be high. The PM_{aged} emissions were dominated by the organic fraction.

Among the precursor gases, NMOGs exhibited notably high emission levels, exceeding those observed in all other transport sectors studied.

NO_x emissions were in line with values reported in supporting studies. The aviation sector currently lacks effective NO_x mitigation technologies, although some sustainable aviation fuels (SAFs) may offer modest reductions. NH_3 emissions were low, as expected, since aircraft engines do not employ EAT systems inducing NH_3.

Due to the high dilution ratio in the measurement setup in aviation simulation, SO_2 emissions could not be quantified, while SO_2 levels were calculated based on the sulfur contents of the aviation fuels used.

Experiments with permeation oven

The influence of VOCs on SecA formation in the presence of exhaust emissions was studied in the laboratory marine exhaust simulation campaign. Preliminary permeation oven results indicate that, as expected, most aromatic compounds seemed to contribute to SOA formation, while no clear increase was observed in the case of dodecane. The extent of SOA formation also varied depending on which individual compound was added to the exhaust mixture. For instance, the addition of p-xylene did not appear to lead to a noticeable increase in organic aerosol concentrations, whereas benzene seemed to cause a more evident enhancement. Naphthalene also appeared to contribute to some increase in SOA, although the effect was more moderate. Interestingly, results were also different depending on the type of fuel used. These differences may be related to the compounds' varying SOA formation potentials, atmospheric lifetimes, emission ratios, and chemical behavior under the experimental conditions

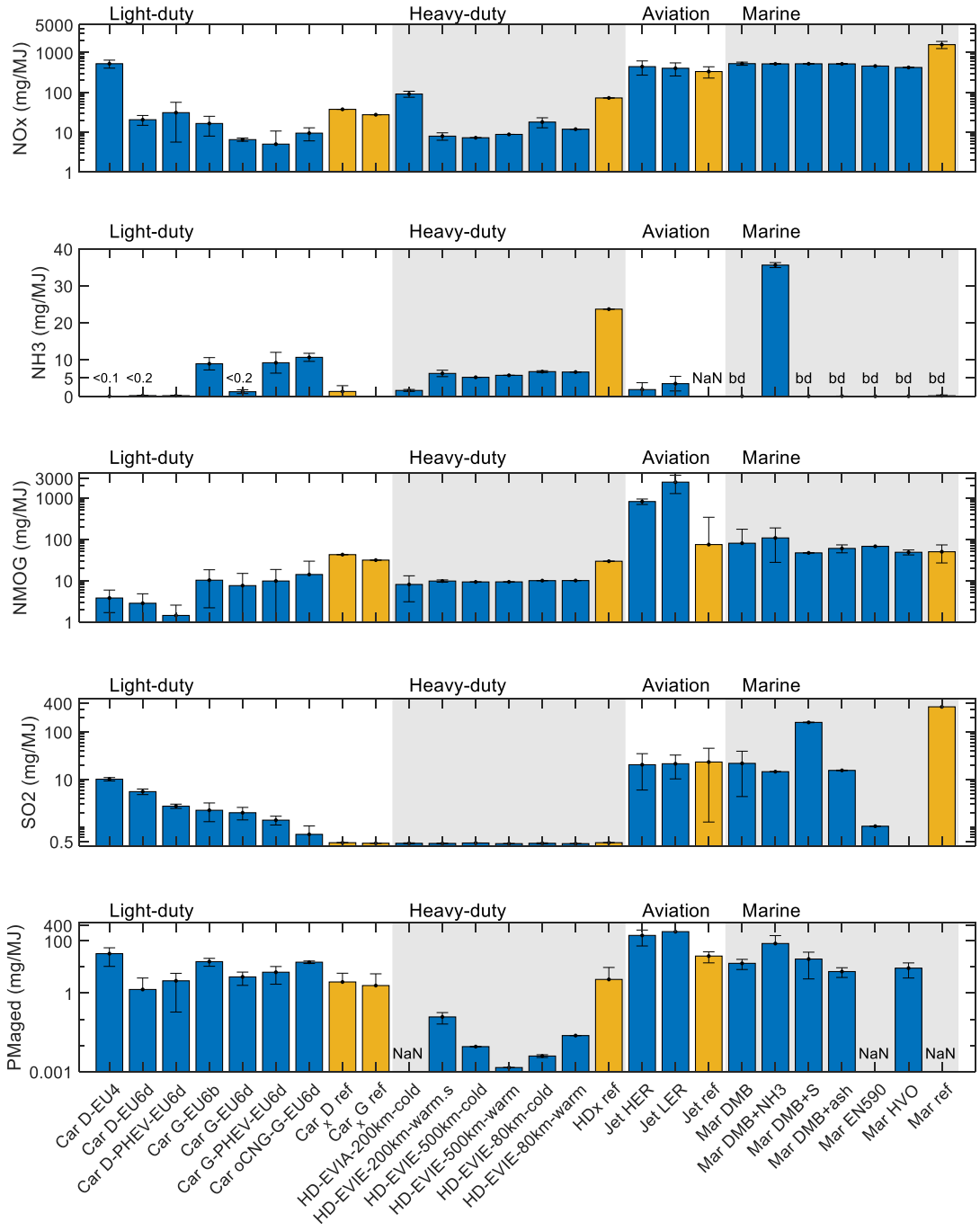


Figure 8. Overview of precursor emissions (NOx, NH3, NMOG, SO2) and PMaged measured in PAREMPI as averages of replicate tests at different temperatures and error bars represent deviations (blue bars). Reference values, orange bars, are explained in Appendix 1. Due to large variations in emissions between technologies, logarithmic scales are used.

3.1.3 Composition of PM_{aged} emissions

Figure 9 shows the average contribution of organics, nitrates, sulfates, and ammonium to aged submicron particles in exhaust emissions for different sectors measured with the SP-AMS. Large

variation in the composition of emissions was seen depending on engine, aftertreatment and fuels. For cars, the emissions were dominated either by organics or nitrate. Ammonium was clearly present in SecA for most vehicles. However, the role of NH_3 in exhaust was particularly important in promoting formation of nitrates, a phenomenon also observed in marine exhaust emissions. However, we note that during the driving cycle, variation in composition of PM was significant (Simon et al., 2025) and especially the cold start affected composition of emissions. For HD vehicles, the emissions were dominated by either organics or nitrate, similarly to cars. Also, the cold start had a major impact.

For marine emissions, the fuel had significant influence on emissions. Sulfur content of fuel had clear impact on sulfate emissions, and added NH_3 clearly impacted nitrate and ammonia emissions. For other marine fuels, the emissions were dominated by organics, with minor amounts of inorganic ions. For aviation, the emissions were dominated by organic compounds with very small contribution of inorganic ions.

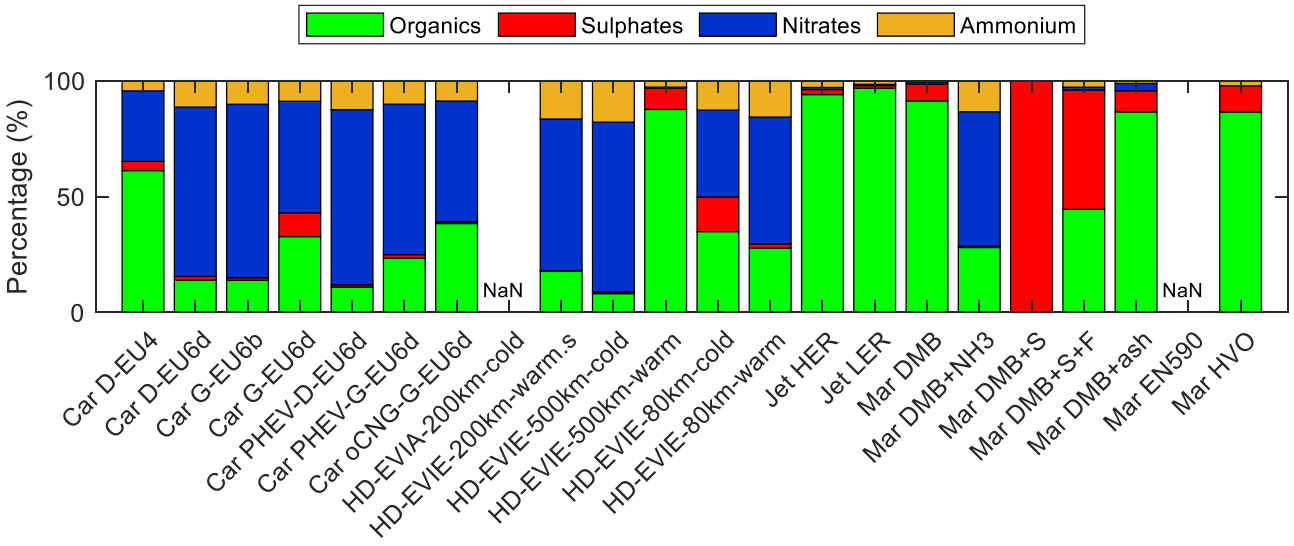


Figure 9. Composition of PM_{aged} measured with the SP-AMS in the PAREMPI measurements for different sectors.

3.1.4 Ultrafine particle emissions

Traffic is an important source of ultrafine particles (UFP with diameter <100 nm, e.g. Harni et al., 2024, Kuittinen et al., 2020) with significant concentrations of particles down to few nanometer size range (Rönkkö et al., 2017). In the PAREMPI the number concentration of particles was measured in different size ranges (e.g. D_p > 1.5nm, 10nm, or 23nm). Particle number size distribution was measured in several experiments and, in addition, the non-volatile fraction of PN was measured separately for $PN_{nv}10$ and $PN_{nv}23$ fraction.

Results, reported more comprehensively in PAREMPI deliverable **D2.2** and in upcoming articles, indicate that exhaust filtration significantly reduces the fresh exhaust ultrafine particle emissions. E.g. in the HD experiments particle number concentrations of fresh exhaust were very low. These very low emission levels can cause challenges e.g. for the accuracy of measurements as well as need for further development of exhaust sampling and more sensitive particle measurement devices. Regarding LD cars, PAREMPI results show that exhaust contains particles also in particle sizes smaller than 10 nm. Emissions of these small particles are not regulated. However, sub-10 nm particle emissions of traffic have been observed to significantly affect urban air particle number

concentrations in traffic influenced areas (Rönkkö et al., 2017). In addition to road traffic, recent studies on aviation impacts on urban air quality have shown surprisingly high role of ultrafine particles from airport activities (e.g. Lepistö et al., 2026). This indicates that the representativeness of the current particle number-based emission standards should be considered carefully, especially related to the particle size range covered and gas-to-particle processes that can impact especially on fresh exhaust particle number and thus on air quality near emission sources.

In the particle number results above, the reported values include both volatile and non-volatile particles, while current emission standards consider only non-volatile particle number emissions. Measured non-volatile particle number emissions for particles larger than 10 nm are shown in Figure 10, highlighting the effectiveness of exhaust aftertreatment, especially particle filtration in modern cars and heavy-duty vehicles. In contrast, exhaust aftertreatment technologies were not used in simulations conducted with jet and marine fuels.

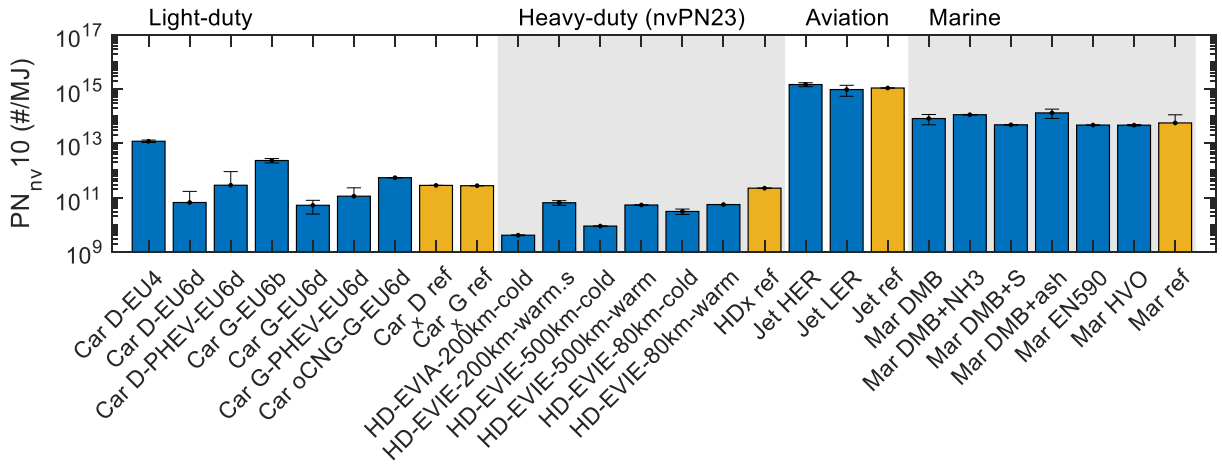


Figure 10. Measured number emission (#/MJ) of PN$_{nv}$10 for different transport sectors (PN$_{nv}$23 for heavy-duty trucks). Reference values, ***orange bars,*** *are explained in Appendix 1.*

3.1.5 Semi-volatile compound and PAH emissions

Quantifying and characterizing semi-volatile compounds (SVCs) in exhaust from transport applications remains challenging (Section 1.2). Compared to particulate matter, the SVC fraction is less known. The semi-volatile compounds, such as PAHs, can be present in either the gas or particle phase, depending on sampling conditions, especially temperature and pressure. Even the tight exhaust emission standards in the road transport sector do not address PAHs, nitrated PAHs, oxygenated PAHs, or dibenzothiophenes (DBTs), and particularly not in the SVC fraction of exhaust.

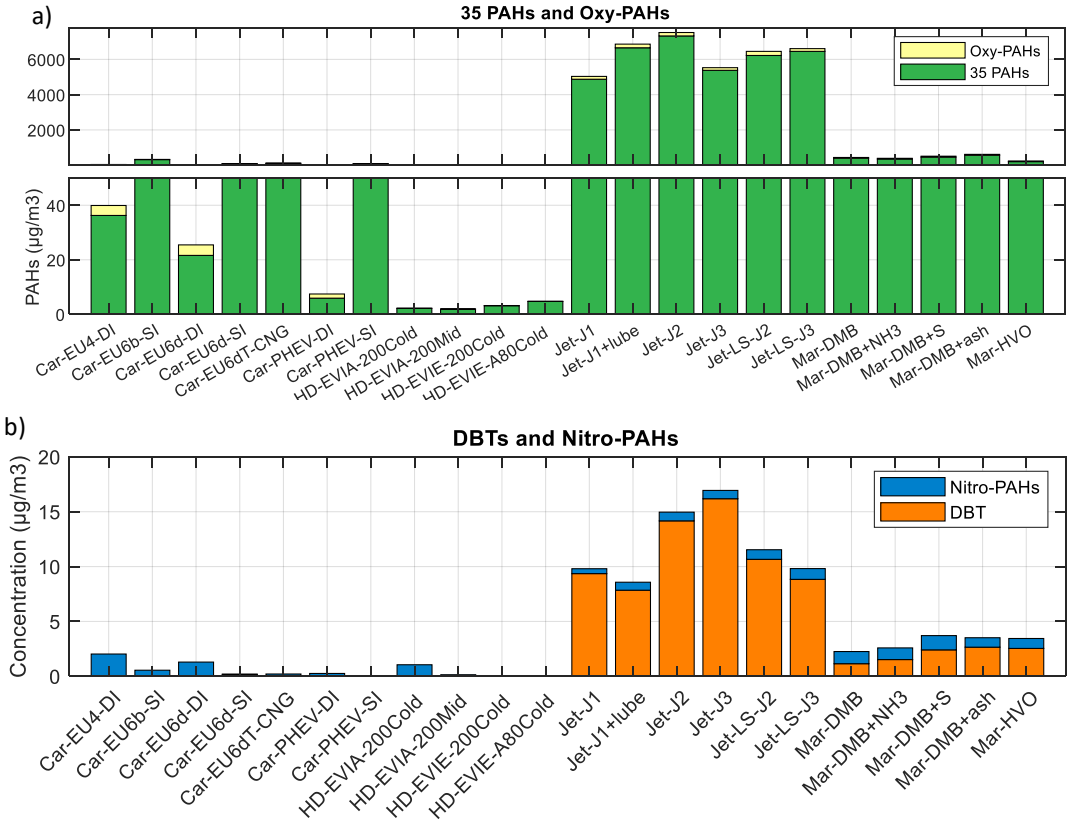
In PAREMPI, both PM and SVC sampling were conducted across all emission measurement campaigns to capture this underrepresented fraction of exhaust emissions. Concentrations of PAHs, nitro-PAHs, oxy-PAHs, and DBTs, summed over PM and SVC fractions, are presented in Figures 11a–c. List of analyzed PAHs can be found from Appendix 2. PAHs were generally more abundant in the SVC fraction than in PM (Figure 11c), although some PAH derivatives were found primarily in PM. Heavier, more carcinogenic PAHs, were predominantly present in PM, while lighter (2-ring) PAHs dominated the SVC fraction.

The results for cars and trucks represent real-driving conditions at cold ambient temperatures. For all spark-ignition (SI) cars, observed PAH concentrations were higher than those for diesel cars or

trucks. Conversely, diesel cars exhibited higher concentrations of oxy- and nitro-PAHs compared to SI cars. Oxy-PAHs were detected in exhaust even for the most modern diesel vehicles, while nitro-PAHs were observed in an older truck. DBT concentrations were generally low for both cars and trucks. Notably, even the most modern vehicles emitted PAHs and in some cases their derivatives (oxy-PAHs, nitro-PAHs, and DBTs).

Aircraft emission simulations revealed extremely high PAH concentrations in exhaust, especially light 2-ring PAHs, along with elevated oxy-PAH and DBT levels. Fuel-related differences were observed, but trends were inconsistent; for example, fuels showed reversed PAH levels under different CAST settings. Marine diesel engine simulations produced PAH concentrations higher than those from cars and trucks, but lower than those from aircraft simulations. Similarly to aviation, fuel-related differences in PAH emissions for marine engines may not be significant. Nevertheless, levels of PAHs, oxy-PAHs, and DBTs in both aviation and marine sectors warrant further investigation.

PAH emissions, primarily originating from incomplete combustion of fuel and lubricating oil, pose serious health risks. These findings underscore the need for continued development of fuels, lubricants, engines, and aftertreatment technologies to mitigate such emissions, as well as experiments with real engines to verify the observations in real-scale settings. Many PAH groups detected in this study are not currently included in air quality monitoring programs.



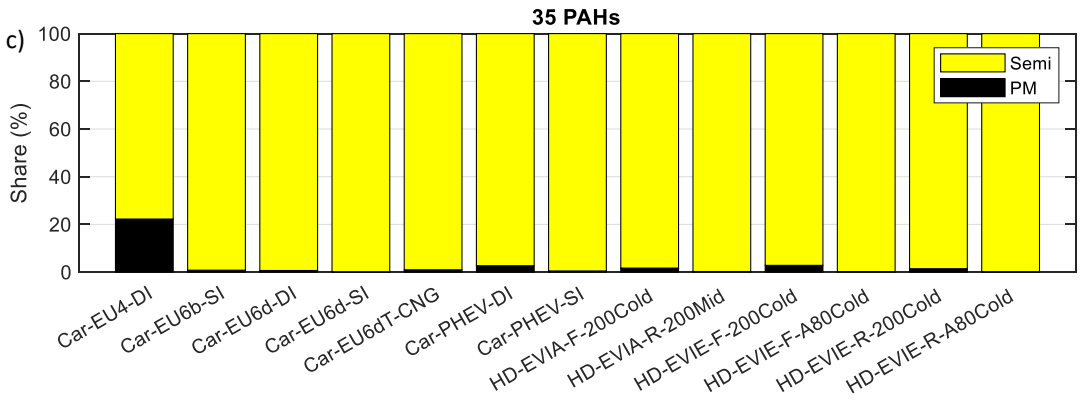


Figure 11. Concentrations of a) PAH and oxy-PAH b) nitro-PAH and DBTs in particle and semi-volatile phases of exhaust measured in PAREMPI campaigns. c) Distribution of PAHs between PM and semi-volatile phases.

The SVC fraction contains also other compounds than PAHs, therefore additional screening methods were evaluated. PM and SVC samples collected from a Euro 2 diesel car were analyzed using thermal desorption gas chromatography–mass spectrometry (TD-GC/MS) and the total carbon method. TD-GC/MS revealed that the SVC fraction contained light oxygenates, aliphatic and aromatic hydrocarbons, and tentatively nitrogen-containing compounds. Similarly, GC/MS analysis of methanol (MeOH) extracts identified aliphatic and aromatic hydrocarbons, along with unidentified acids and esters. Previously, SVCs from a heavy-duty diesel engine were found to contain diesel-type hydrocarbons in the C9–C19 range, distilling between 160–375 °C. The total carbon method indicated that the carbon content in the SVC fraction was several times higher than in the PM fraction. Notably, individual compounds such as cyclosiloxanes were detected in the SVC fraction.

3.1.6 Brake wear emissions Brake wear emissions

The non-exhaust emission characterizations were fulfilled with a campaign for break wear emissions (Figure 12). The brake wear studies showed that emissions depend on pad type and braking conditions. About 30-50% of the PM_{10} emissions were within the $PM_{2.5}$ size range. Moderate UFP emissions can occur during harsh braking. BWP were dominated by iron oxides. BWP components absorb in IR region and thus contribute to measured BC from road transport.

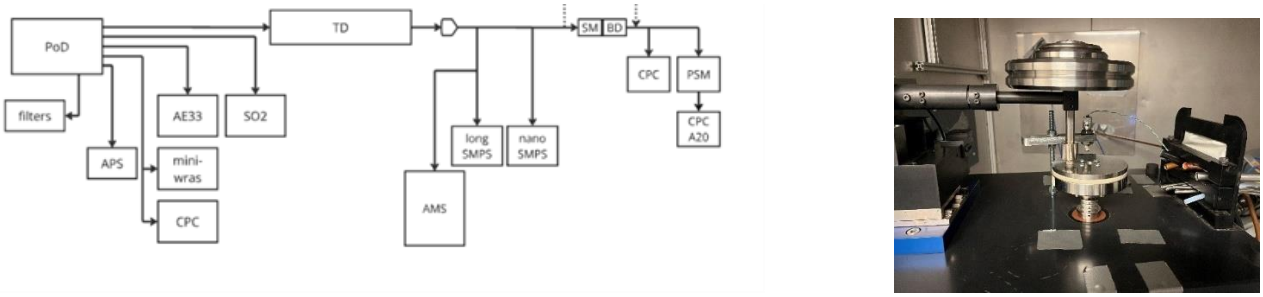


Figure 12. Experimental Set-up used in brake wear measurements (left) and photo of used Pin-on-disk tribometer (Nanovea® T50 tribometer).

3.2 Database on SecA formation potential of emission from transport sectors

The PAREMPI database is described in PAREMPI **D5.1** report and the database as an Excel file is soon available at https://parempi.eu/. PAREMPI **D5.2** report evaluates results of the database. The database was developed as a comprehensive resource to consolidate emission data on secondary aerosol species, their precursors, and associated toxicity from various transportation sources. The database focuses on freshly emitted particulate matter (PM_{fresh}) and PM after ageing (PM_{aged}) through the generation of secondary aerosols. It incorporates direct measurement results from single emission sources such as engines, passenger cars, and heavy-duty vehicles, while excluding mixed emissions like those from roadside or tunnel measurements and data derived from indirect calculations. The database covers both fresh and aged PM and their chemical composition (e.g., organics, nitrate, sulphate, ammonium, BC/EC). It also includes precursor gases and VOCs and SVOCs that may contribute to secondary aerosol formation via atmospheric oxidation reactions (e.g., NO_x, NH_3, SO_2, aromatics). Additionally, the database reports PN and other species such as CO_2 and non-methane organic gases (NMOG). From each study included in the database, detailed information is provided on the classifications of transport sectors (light-duty, heavy-duty, marine and aviation), emission classes, engine combustion technologies, fuel types, engine sizes and measurement conditions, including driving cycles, temperatures, and geographical locations, ensuring a thorough understanding of emissions under various scenarios. Measurement techniques employed in the studies included advanced devices and instruments, which are also documented in the database. **Of particular importance are results of OFRs**, which are currently the most widely used technique for simulating atmospheric aerosol formation in emission studies. The experiments from literature are traceable via references. **Articles from the past 10 years were prioritized**, but older articles can be also included in the database if more recent data is missing or insufficient. Furthermore, peer-reviewed scientific literature, European Union (EU) reviews, or published/unpublished results from previous projects within the Consortium are favored. Presented in an Excel format, the database is easily accessible and can be updated with latest results from PAREMPI project and relevant literature.

The first database version is literature-based only and includes two sections 1) SecA emissions from different transport sectors and 2) Toxicological impact of different transport sectors (see sections 3.2 and 5.1.3). The final database will include also results from PAREMPI measurements, and all literature data and results will be converted in per MJ unit.

Literature data on secondary aerosol emissions from transport sources appeared to be sparse for cars and heavy-duty vehicles, and especially so for ships, aircrafts, and motorcycles. Table 3 summarizes the current status of knowledge on SecA and key contributions made in PAREMPI. Initial findings, based solely on literature, showed limited data for heavy-duty vehicles, ships, and aircraft, with only few real-world, real-driving, or on-board studies available. PAREMPI's own measurements will partly fill these gaps. Early results highlighted that older diesel cars without diesel particulate filters (DPFs) emit significantly more particles than modern DPF-equipped vehicles. Gasoline direct injection (GDI) engines and fuel type were found to influence secondary aerosol formation. Notably, GDI engines also exhibit significant particle emissions, particularly under high-load conditions, as observed in the database. Some challenges identified included the diversity of parameters influencing emission factors (such as emission class, combustion technology, temperature, and fuel), incomplete descriptions of experimental conditions, emission factors reported for only a limited number of chemical species, inconsistent presentation formats (not always tabulated), use of varied units (e.g. mg/kg_{fuel} or mg/km), restricted access to some articles due to paywalls, and the often poor reporting of technical details on transport applications, fuels used, and relevant emissions.

Datasets included in the 1st version of database are presented in Table 3. From literature, the most extensive secondary aerosol dataset was found for light-duty vehicles, comprising 79 data points. For heavy-duty applications, only 28 datapoints met the defined criteria. For aviation, only 3 studies were identified from literature, and none for marine sector. Already before the detailed analysis, and even when considering the oncoming updates from the PAREMPI Consortium, secondary aerosols from marine and aviation sectors warrants further studies.

Table 3. The current status of knowledge and key contributions made in PAREMPI.

Sector	What Is Known from Literature	Key Findings from PAREMPI	Measurements	Knowledge Gaps / Notes
Light-duty	Most extensive literature data among sectors (79 datapoints); aged PM and secondary aerosol formation relatively well studied. Missing new fuels and techs.	PM_{aged} data from 7 modern cars (gasoline, diesel, CNG, PHEVs) at 3 temperatures (-9, 23 and 35°C). High comparability with RDE testing.	Controlled lab conditions simulating RDE; 3 temperatures	Lack of real-world on-road data. FFVs with latest standards are of interest; challenges in applying portable systems.
Heavy-duty	Moderate literature data (28 datapoints), mostly lab-based; limited info on alternative fuels and non-road applications.	New data from 2 trucks (Euro VI-E, VI-A); includes SecA formation in winter on-road tests in northern Finland.	Lab and on-road (cold/dark winter conditions)	Limited data on on-road, off-road and alternative fuels; system suitable for HD applications.
Aviation	Only three published studies reported secondary aerosol emissions (OFR) from aircraft prior to PAREMPI.	New dataset with various jet fuels including SAF blends; significantly expands knowledge.	Simulated aircraft emissions	Still limited data; further research needed.
Marine	One published study from on-board conditions	Studies with diesel engines using marine fuels varying SO_2, NH_3 and ash concentrations in exhaust.	Simulated marine emissions	Field is largely unexplored; database updates will clarify marine engine roles in secondary aerosol formation.

The database, in its final form, will be versatile, supporting research, policy-making, and engineering applications by enabling a holistic assessment of transport emissions and their environmental and health impacts. Its comprehensive, user-friendly structure makes it a valuable tool for evaluating and mitigating the role of transportation in air quality degradation and health impacts of novel transport technologies.

3.3 Identification of gaps in knowledge and discussion

1. Sectoral and Technological Coverage of SecA results

There is a notable lack of data across various transport sectors and technologies:

- Aviation SecA data is very sparce in literature and marine SecA emission data almost completely missing, especially on-board, in-field measurements
- Non-road mobile machinery, motorcycles and non-exhaust emissions (e.g., brake and tire wear and tire-road wear) are underrepresented in current datasets.
- Heavy-duty vehicles have been studied far less than light-duty vehicles, especially under real-world conditions.
- Emerging fuels (e.g., methanol, e-fuels) and new engine and after-treatment systems (EATS) are not characterized in terms of their secondary emission profiles.
- PM_{aged} measurements in Real Driving Emission (RDE) conditions, except PAREMPI truck measurements, remain almost non-existing, despite their importance in capturing realistic emission behavior.

2. Comparability of Experimental Results

A major challenge in synthesizing findings across studies is the lack of standardization in experimental setups. Differences in dilution, chamber design, aging protocols, and analytical methods hinder the comparability of PM_{aged} emission results, making it difficult to draw consistent conclusions. Furthermore, the results are given in variable units (mg/m^3, mg/kg_{fuel}, mg/km, mg/MJ), which further complicates the comparison without conversion to harmonized unit. In PAREMPI, results from literature and from PAREMPI measurements will be converted to harmonized unit (per MJ) to increase transparency of PM_{aged} results across transport sectors.

4 Modelling of SecA and PM_{aged} emissions

4.1 Description of ePMI module

In PAREMPI we are developing a novel digital equivalent total particles emissions index module (ePMI) software. The ePMI represents primary and secondary aerosol particle number and mass formation from anthropogenic sources, with focus on the transport sectors. The ePMI is intended to be used as a computationally efficient module for global and regionals scale chemistry transport models. We are currently working on the implementation of ePMI in the global chemistry transport model TM5. The ePMI module includes simplified parameterizations of biogenic and anthropogenic SOA and UFP formation rates based on a limited number of input variables ([NO_x], [O_3], [OH], [H_2SO_4], VOC mixtures, [NH_3], temperature *etc*), which are standard variables provided by regional and global scale CTM. The ePMI module is based on the existing and widely used modal aerosol dynamics module (M7) (Vignati et al., 2004; van Noije et al., 2014), improved anthropogenic SOA volatility basis set (VBS) parameterizations developed in PAREMPI, and novel NH_3-H_2SO_4 NPF lookup tables based on the Atmospheric Cluster Dynamics Code (ACDC, Olenius et al., 2013). We have implemented and tested the NPF lookup table together with M7 in the global chemistry transport model TM5 in EC-Earth3 (Svenhag et al., 2024, 2025), and we are currently working on the evaluation and implementation of simplified representations of anthropogenic SOA VBS schemes, analogous to the existing biogenic SOA formation schemes used in EC-Earth/TM5 (Bergman et al., 2022). Similar to the NPF parameterizations, the intention is that the ePMI VBS scheme will be in the form of a lookup table which dynamically account for how the complex atmospheric chemistry (RO_2, NO_x, OH etc.) and temperature influences the OVOC volatility distribution and particle size resolved SOA growth rates.

4.2 Transport emissions impact on the air quality in Europe

Figure 13 shows the compiled modelled yearly average UFP and PN (D_p>100 nm) concentrations in the atmospheric surface layer over Europe in 2018. The impact from ship emissions on the UFP concentrations along the major ship lanes are clearly apparent in the Adriatic Sea, Mediterranean Sea, the North Atlantic Ocean, English Channel, North Sea and the Baltic Sea, with major impact on the UFP levels over the ocean and coastal regions in Europe. For particles larger than 100 nm in diameter the geographical number concentration patterns look markedly different, with hotspot regions in the Po Valley in Northern Italy and East Europe, in particularly over the Balkan countries.

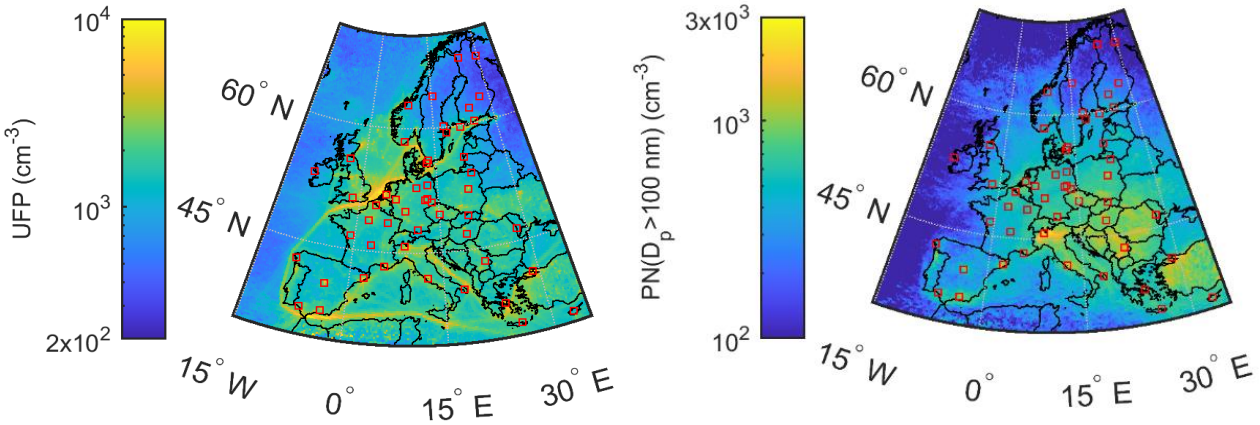


Figure 13. Modelled (ADCHEM) average surface concentrations of UFP (left panel) and particles >100 nm in diameter (right panel) in Europe in year 2018.

To investigate how different emission sectors and processes (e.g. NPF) influence the particle concentrations in different regions in Europe we performed several ADCHEM sensitivity model runs. Considering the scope of the PAREMPI project, the most extensive sensitivity simulations, covering all regions over the European continent, were performed to assess how road traffic emissions influence the air quality. According to our ADCHEM model simulations the road traffic in Europe has major impact on the nitrogen oxides ($NO_x \equiv NO + NO_2$) levels and indirectly the ozone and PM levels (nitrate aerosol mass) in central Europe and urban regions in general (Figure 14).

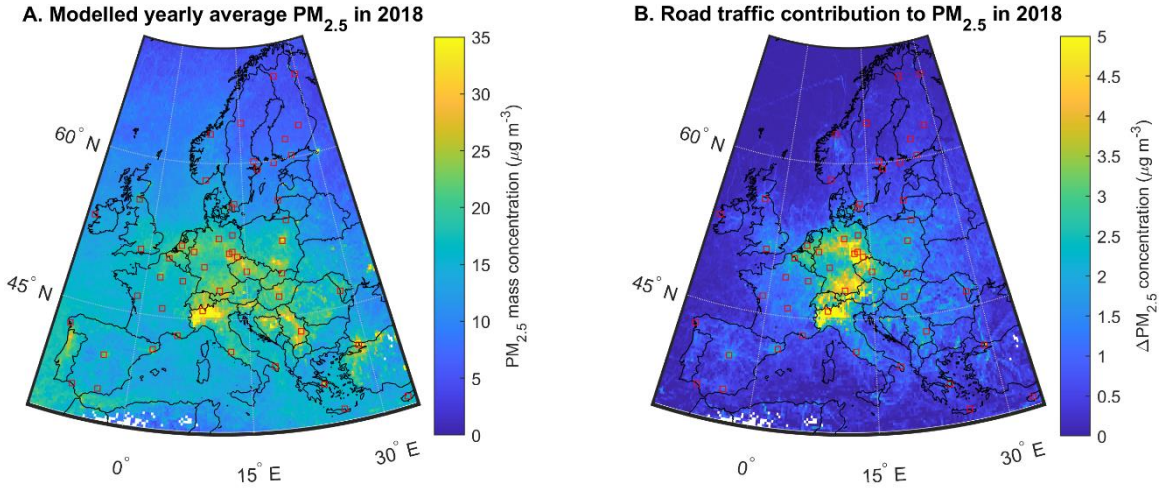


Figure 14. Modelled average $PM_{2.5}$ levels over Europe in the year 2018. Left panel shows the absolute concentrations from the basecase model simulations including emissions from all known sources. Right panel shows the concentration difference between the basecase and simulations with road traffic emissions turned off in the model (WoRoad).

According to our model simulations, most of the $PM_{2.5}$ mass from road traffic is nitrate PM (Figure 15) formed as SIA from the road traffic NO_x emissions. A smaller share of the road traffic emissions contribution to the $PM_{2.5}$ levels in Europe are BC, POA and Non-exhaust emissions (see PAREMPI **D3.3** report). According to our model simulations, the road traffic emissions have relatively minor influence on the SOA mass concentrations in Europe and even a minor net negative contribution to the SOA mass in regions with intense road traffic. The main reason for this is that the road traffic has high NO_x and relatively low and slowly oxidising VOC emissions which results in delayed SOA formation. The NO_x emissions results in complex non-linear effects on the atmospheric chemistry that governs not only the oxidation of the emitted anthropogenic VOCs, but also the oxidation of biogenic VOCs.

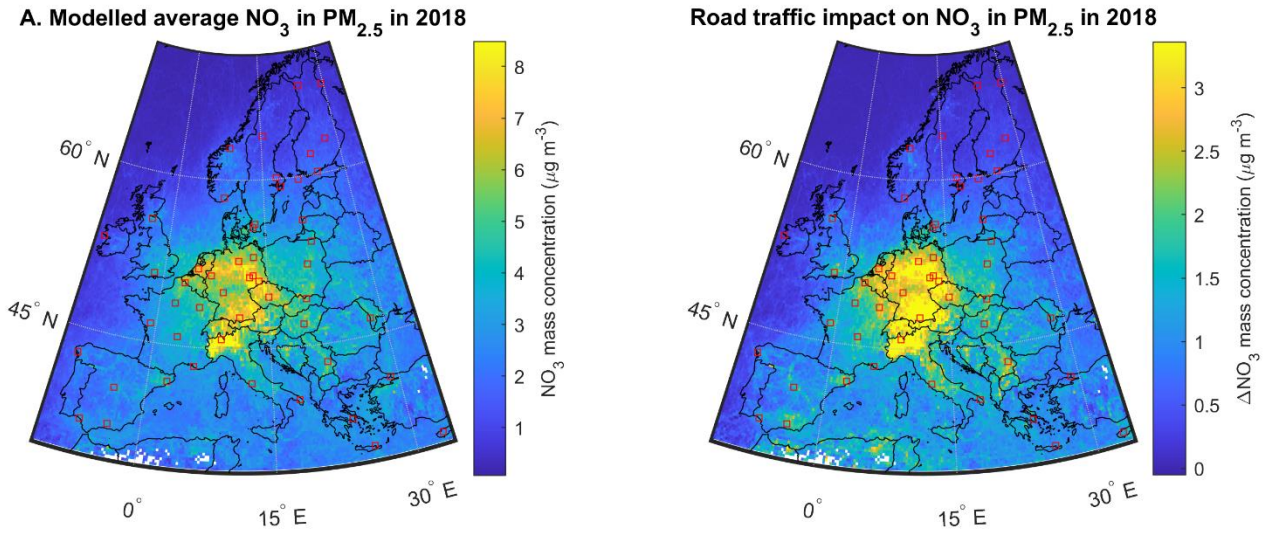


Figure 15. Modelled average nitrate mass contribution to the $PM_{2.5}$ levels in Europe in the year 2018. Left panel shows the absolute concentrations from the Basecase model simulations. Right panel shows the concentration difference between the Basecase and simulations with road traffic emissions turned off in the model (WoRoad).

4.3 Modelling of atmospheric SecA formation

The road traffic emissions in Europe cause complex non-linear effects on atmospheric chemistry and secondary aerosol formation. Road traffic NO_x emissions in combination with agriculture NH_3 emissions are an important source of SecA $PM_{2.5}$ in the form of ammonium nitrate. According to our European scale model simulations, NH_4NO_3 is the dominating component of $PM_{2.5}$ in most regions of Europe during the winter and early spring. The total OA $PM_{2.5}$ content shows less variation over the year, but POA is the dominating OA source in the wintertime while SOA dominates during the summer season. NO_x emissions have a major impact on the VOC oxidation chemistry and tend to suppress the SOA formation. Thus, although the road traffic contributes to important SOA precursors (aromatics and large alkanes) the net impact on the atmospheric SOA levels can be both positive and negative depending on the amount of BVOC emissions. The substantial NO_x emissions are also an important source of tropospheric ozone and increase the atmospheric hydroxy radical (OH) concentrations (the atmospheric oxidative capacity), especially during extreme ozone episodes during European heatwaves with high BVOC emissions and NO_x-limited ozone production (Figure 16). Thus, preventing road traffic NOx emissions, especially during forecasted heatwaves, can be an efficient measure to mitigate dangerous tropospheric ozone levels in Europe.

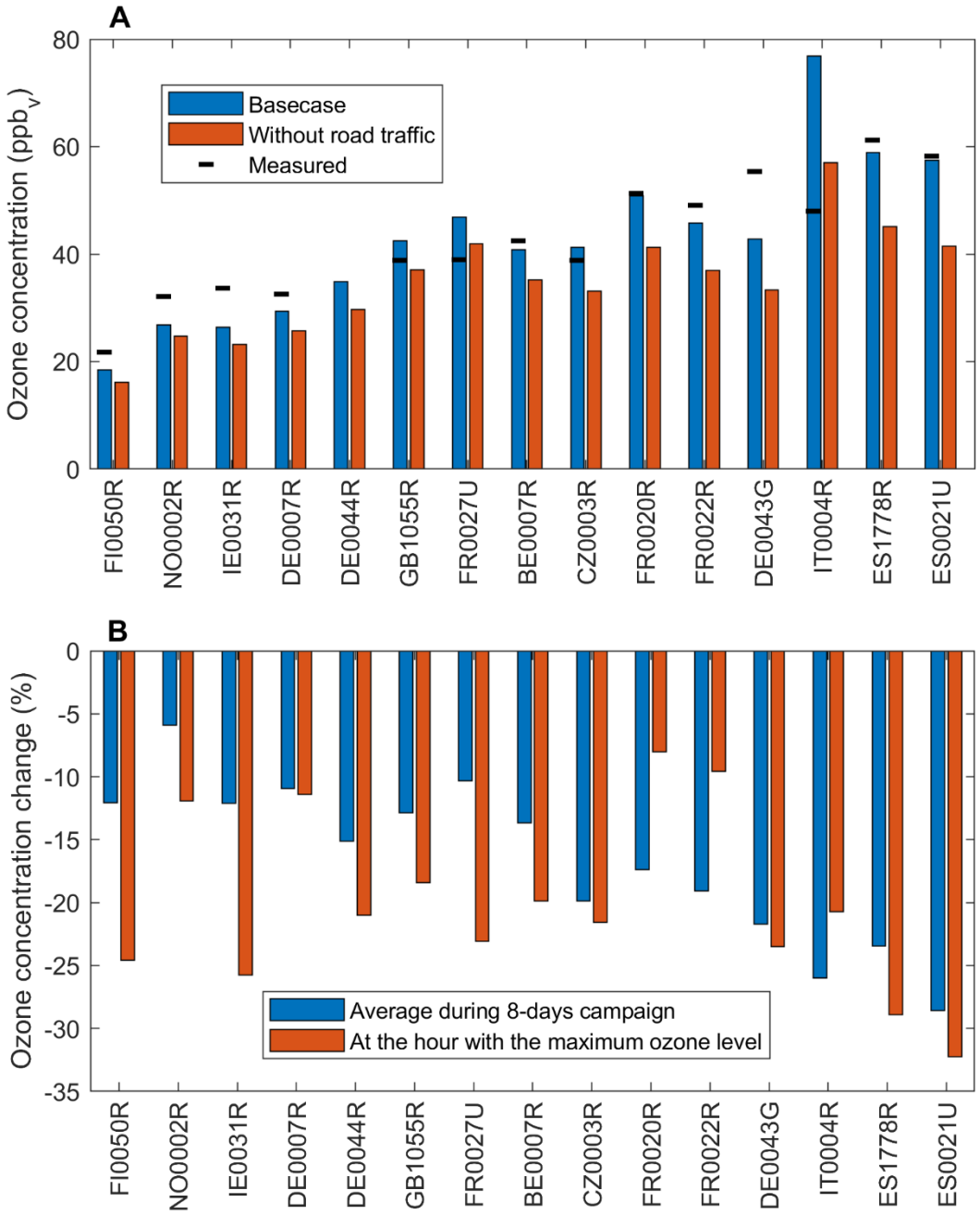


Figure 16. Measured and modelled (ADCHEM) average surface ozone concentrations (panel A) and relative ozone concentration changes as a result of the hypothetical removal of road traffic emissions in Europe (panel B). The results are from a European summer heatwave episode from July 12-19, 2022, when over 30 stations participated in an intensive EMEP VOCs, ozone and carbonaceous aerosols measurement campaign (EMEP, 2023). Results from 15 of these EMEP stations are show in the figure. Panel B shows both the modelled average and maximum peak ozone concentration decrease which would be achieved if all road traffic exhaust emissions were removed in Europe.

4.4 Identification of gaps in knowledge and discussion

During the PAREMPI project we have made major advances in understanding the oxidation chemistry, including RO_2 autoxidation, that leads to SOA formation from major aromatic ASOA precursors (Iyer et al., 2023; Pichelstorfer et al., 2024, PAREMPI **D3.2** report). However, for the majority of the AVOCs, including many alkanes and PAHs, that we have identified during the vehicle exhaust campaigns in PAREMPI (see Sect. 3.1.) we still lack the realistic gas-phase oxidation mechanisms that produce condensable organic compounds leading to SOA formation at realistic atmospheric conditions, including variable NO_x and temperature conditions.

5 Health effects of transport emissions

5.1 Toxicity evaluation

5.1.1 Description of newly developed toxicity incubator and measurements

Within PAREMPI, an existing portable air-liquid interface (ALI) exposure system, referred to as the "toxicity incubator," was extended for campaign-specific applications. This system is described in robust measurement system report PAREMPI **D2.3.** The system enables direct exposure of human respiratory cells to diluted exhaust emissions in both mobile and stationary settings (Figure 17).

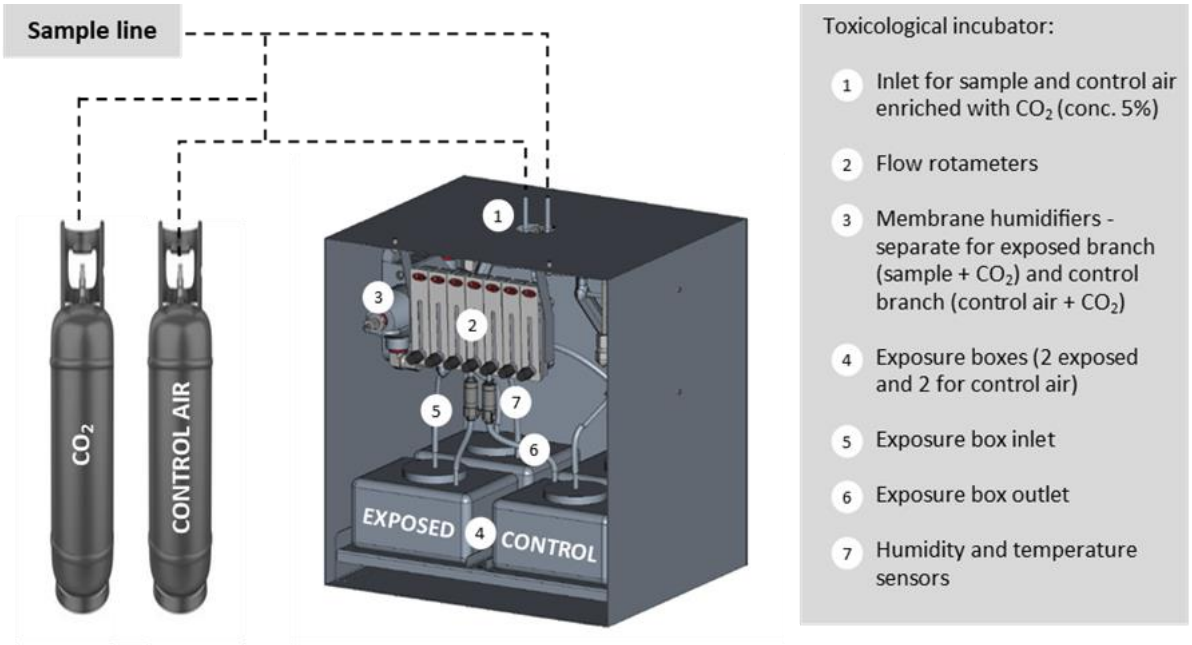


Figure 17. Toxicological incubator used during PAREMPI campaigns. Detailed information is presented in Vojtisek-Lom et al., (2025).

Toxicological incubator used during PAREMPI campaigns is presented in detail in the article by Vojtisek-Lom et al., (2025). The exposure unit consists of airtight stainless-steel boxes (17 × 13 × 9 cm), each accommodating eight 6 mm Transworld inserts in a 24-well plate configuration (Figure 18). These boxes are designed to be gas-tight and allow even distribution of aerosols over the cell

surface. They are connected via inlet and outlet ports to controlled air supply lines, enabling exposure to either filtered control air or diluted exhaust. Each exposure box is placed within a compact incubator (40 × 35 × 45 cm), which ensures stable environmental conditions: 5% CO_2, 37°C temperature, and >85% relative humidity. The incubator is thermostatically controlled and equipped with internal fans to maintain uniform conditions. Conditioned air—either clean or containing diluted emissions—is drawn through 2–4 exposure boxes simultaneously, at a flow rate of 25 cm³/min per insert. This setup supports parallel exposure of control and test samples under identical thermal and humidity conditions. Deposition tests showed ~50% particle loss at 15 nm and ~1.5% deposition efficiency for 10–21 nm particles. The system operated across −7°C to +32°C.

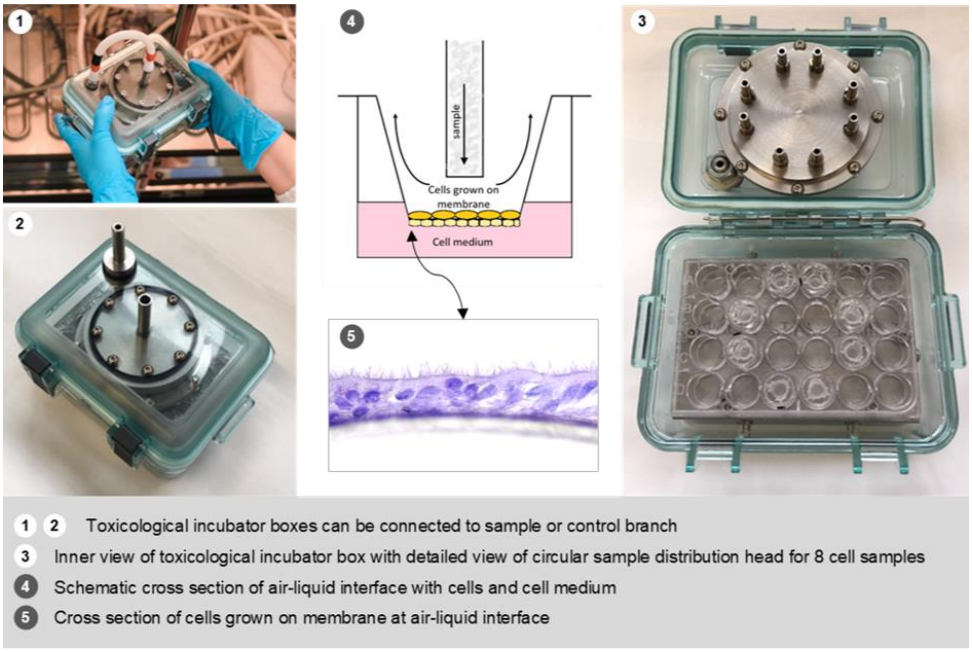


Figure 18. Air-liquid interface system for cell exposure as used in Vojtisek-Lom et al. 2025

In all PAREMPI campaigns, BEAS-2B and MucilAir™ cell models were exposed to both PM_{fresh} and PM_{aged} generated using oxidation flow reactor (OFR). Exposure durations were 1–2 hours. To replicate lung-relevant conditions, no artificial deposition enhancers (e.g. charging or thermophoresis) were used. An ozone trap was placed upstream of the OFR to eliminate residual ozone and avoid artefacts.

Campaign-specific setups used in PAREMPI measurement campaigns:

- **Light-duty campaign:** Exposures to car exhaust were performed next to the chassis dynamometer test cell at BOSMAL. The exposure chamber was connected via Tygon tubing and the dilution ration was: avg. DR (23°C) = 49; avg. DR (-9°C) = 58. Exposure time was 72 min per day (each sample was exposed twice, in total 144 min exposure/sample).
- **Marine and Aviation** campaign: Conducted at VTT, the experiments used a diesel generator running on marine fuel to mimic exhaust formation from a marine engine, and a CAST generator to simulate aircraft exhaust (see section 2.2). The exposure chamber was connected to a PAM reactor via Tygon tubing. The dilution setup included the following components: an E-diluter DR=5:1 or 10:1 depending on cell model complexity; a PTD DR=15.4 for marine and DR=13.5 for aviation; an ejector diluter with DR=4:1; and a pre-PAM dilution of DR=1.7. Exposures were performed on both complex 3D models and simpler BEAS-2B cells. The 3D models were exposed with the PAM UV lights turned off, whereas in the BEAS-2B experiments the PAM UV-lights were alternated on and off (fresh and aged). Ozone was removed from the sample prior to BEAS-2B exposures using an ambient-temperature catalytic stripper, because ozone is

generated in the OFR and is harmful to the cells even at very low levels. The ambient temperature catalytic stripper had only modest effects on the particles and VOCs. Exposure time was 90 min (E-diluter 10:1 for aviation, 5:1 for marine). For 3D models (PAM off), the stripper was bypassed and exposure extended to 120 min using a 5:1 E-diluter.

- **Heavy-duty campaign:** The ALI system was mounted inside a trailer towed by HD trucks, enabling mobile, real-driving exposure sampling in Tampere. The exposure chamber was connected via Tygon tubing and the dilution was set for Truck 1 (DR_{avg}=48) and for Truck 2 (DRavg=65). The exposure time was according to the route Tampere-Jämsä-Tampere (avg. 180 minutes for Truck 1 measurements and 240+ minutes for Truck 2 with defect).

5.1.2 Toxicity results from PAREMPI

Toxicological results from the PAREMPI campaigns show that while cytotoxicity was low, clear molecular-level effects—such as oxidative stress, DNA damage, and gene expression changes—were observed. These findings highlight the need for in-depth analysis and also address the unregulated pollutants like VOCs, sub-10 nm particles, and PAHs, which contribute to health impacts beyond current emission limits.

The integration of the air–liquid interface (ALI) exposure system into real-world testing was a key advancement, enabling direct in situ toxicity assessment. However, these setups are complex, resource-intensive, and inflexible, making the testing challenging and sometimes not fully successful.

From a prevention perspective, toxicological insight should be included in early design of engines, fuels, and aftertreatment systems. While cheaper early screening helps avoid high-risk choices, full impacts often result from system-level interactions, real-world testing (including SecA formation, and multi-endpoint biological analysis, environment) is needed to capture toxicological effects. To enable this, measurement campaigns must be planned to support toxicological studies through appropriate exposure conditions and sample handling and scheduling.

Preliminary toxicity findings are presented in PAREMPI **D4.2**. In this summary, these results are qualitatively assessed (Table 4). Exhaust emissions varied across measurement campaigns involving cars, heavy-duty trucks, marine engine and aviation turbine simulations, influenced by technologies, fuel properties, and operational conditions. For example, organic compounds were predominant in the exhaust from an older diesel vehicle, jet fuel combustion, and during operation with distillate marine fuel (DMB). In contrast, PM_{aged} was primarily composed of nitrate and ammonium species under conditions with elevated NH_3 levels were injected in the exhaust (DMB + NH_3), while sulfate species dominated with high-sulfur marine fuel (DMB + S).

Despite the compositional differences, overall exhaust toxicity remained low. Trans-epithelial electrical resistance (TEER), a strong indicator of cell barrier integrity and permeability, was largely preserved, and oxidative stress responses were minimal. However, transcriptomic profiling revealed changes in gene expression, indicating that certain biological pathways were responsive to the exhaust exposures.

Table 4. PM_{aged}, sum of 35 PAHs (section 3.1.5) and toxicity ratings in PAREMPI campaigns evaluated preliminarily, and will be more detailed in oncoming publications. The four key toxicity endpoints (C, G, O, D) are described in section 5.1.3. For toxicity, exhaust volumes for cell exposures were smaller in aviation than in other campaigns reducing comparability between transport sectors. Color codes from white to dark red indicate rating from not significant to very significant adverse effect.

Sector	Technology	Composition		Toxicity							
		Dominating species	PM_{aged} 35 PAHS	Fresh Complex/Simple (BEAS-2B)				SecA Simple (BEAS-2B)			
				C	G	O	D	C	G	O	D
Cars	D-EU4	Organics						-	-	-	-
	D-EU6d	NO3, NH4						-	-	-	-
	G-EU6b	NO3, NH4						-	-	-	-
	G-EU6d	NO_3, Org.						-	-	-	-
	D-PHEV	NO3, NH4		-	-	-	-	-	-	-	-
	G-PHEV	NO3						-	-	-	-
	CNG-EU6d	NO3, Org.		-	-	-	-	-	-	-	-
Trucks	EU VI E, F1	NO3, NH4						-	-	-	-
	EU VI E, F2							-	-	-	-
	EU VI A, F1							-	-	-	-
Aviation	1	Organics						-	-	-	-
	2	Organics						-	-	-	-
	3	Organics									
	4	Organics									
Marine	DMB	Organics									
	DMB+NH3	NO3, NH4									
	DMB+ash	Organics						-	-	-	-
	DMB+S	SO4		-	-	-	-	-	-	-	-
	HVO	Organics		-	-	-	-	-	-	-	-

The primary factor influencing the observed toxicity responses was the cold ambient temperature during the car measurements. Across all measurement campaigns, no consistent relationship was identified between toxicity results and specific engine technologies or fuel types, despite the substantial variability in exhaust chemical composition. Although clear differences in toxicological effects in technologies were not evident, some remarks can still be made (see **D4.2** for details).

- Car measurements showed increased cytotoxicity and TEER changes particularly for PHEV-G, EU6d-G, and older diesel (EU4-D). An elevated response on gene expression was observed in -9°C, but no hard toxicity, so the cells were actively “fighting” the exposure. Oxidative stress responses were not detected.
- On-road tests with two heavy-duty trucks under winter conditions revealed differences in cytotoxicity and DNA damage. While overall transcriptomic responses were modest, EN590 affected more genes than HVO, suggesting fuel composition to play a role even under low toxicity scenarios. No oxidative stress responses were detected. Despite of low PAH levels emitted by trucks (except NPAHs for truck EU VIA), relevant toxicity responses were observed. Truck emitted substantially after cold-start, while these emissions “diluted” over long routes (80 km and 200 km). Notably, toxicological effects once induced cannot be “diluted” later. Further studies with short toxicity exposures covering cold-starts of heavy-duty vehicles would be interesting.
- In the aviation simulation, acute cytotoxicity was minimal, although the SAF fuel blend triggered transcriptomic changes, suggesting low short-term risk but potential relevance for sensitive populations or longer exposures. Despite gene-level changes, toxicity assays showed minimal acute damage. For the CAST combustion system, operational parameters and combustion

stability may have influenced results, suggesting further studies to confirm toxicity findings with different jet fuels. For this aim, consideration of dilution ratios is suggested as explained below.

- In the marine simulation, exhaust with different marine fuels caused low cytotoxicity but significant transcriptomic alterations. The strongest responses were observed when ammonia was present in the exhaust, indicating potential biological relevance despite low toxicity levels.

In the toxicity measurements, raw exhaust exposure levels need to balance two aspects: reflecting realistic human exposure in urban environments and maintaining conditions that preserve cell viability. Assuming a typical human breathing rate of approximately 8 L/min and a roadside dilution ratio of 1000 in areas of heavy traffic, the estimated exposure to raw exhaust equates to roughly 480 mL per hour. This estimate aligns with findings from PAREMPI **D3.1**, which reports dilution ratios of 600–800 at a distance of 10 meters behind a vehicle. The actual raw exhaust volumes used for cell exposures varied (50-86 mL for vehicles, 23-34 mL for marine and 2-6 mL for aviation), due to the corresponding variation in dilution ratios. This affects only cross-sectoral comparisons, while within each sector, technologies and fuels were tested under consistent dilution conditions, ensuring internal comparability. When scaled with human lung surface area (75 m^2) relative to the cell culture insert area (0.28 cm^2), and considering statistical deposition rates, which are an order of magnitude higher for lungs than for the exposure chamber due to the residence time of the inhaled exhaust, the applied dose per cm^2 of lung surface area corresponds approx. one year of inhalation at a 1000:1 dilution ratio in road and marine campaigns, and about one month for aviation. These values are within the same order of magnitude as potential real-world exposures in busy urban environments. Notably, the highest observed biological response was rated as “notable” (score 3 on a 1–5 scale) in all campaigns, indicating that the exposure levels were appropriate to detect effects without overwhelming the system. Excessive dilution in the aviation campaign affected only the cross-sectoral comparison, while conditions were similar internally in each campaign. The extra diluter in aviation campaign was needed due to the insufficient exhaust output from CAST in relation to needs of the measurement setup.

One approach to harmonize sample exposures would be using CO_2 output as a reference metric for carbonaceous fuels. Conditions that elicit clear toxicological responses from older diesel technologies, known for carcinogenic emissions, could serve as a reference. Standardizing raw exhaust exposure by adjusting dilution ratios would enable reasonable exposure times, particularly when evaluating low-emission technologies.

Our findings suggest that even brief exposures can cause relevant toxicological responses, including emissions from the cleanest technologies, with ambient temperature having an important role. An interesting methodological outcome values usefulness of integrating transcriptomic-based endpoints into emission toxicity assessments, since they may reveal early biological perturbations not captured by conventional assays. With clean technologies, traditional toxicological endpoints may lack sensitivity at relevant exposure concentrations. This is especially challenging when sources are distant or samples are highly diluted, potentially masking biological effects.

5.1.3 Toxicity database and rating system

Toxicology section of the database was built to complement the secondary aerosol database and to enable evaluation of toxicological impacts of different transport technologies, fuels and exhaust aftertreatment systems. Toxicological part includes 85 entries covering various aspects of transport emissions and their toxicity assessments. Toxicity database contains data on Exposure Classes such as on-line exposures at the ALI (the Air-Liquid Interface), PM pre-collected (standard and water-based), for both liquid-liquid (traditional) and the ALI testing. Emission Sources include PM fresh exhaust, PM aged (secondary aerosol), and PM mixed with volcanic ash. The database spans

Transport Sectors, including Light-Duty (LD), Heavy-Duty (HD), and Non-Road Mobile Machinery (NRMM), with Applications covering Cars, Engines, Generators, Buses, and Tractor engines. Engine Combustion Types are categorized into Spark Ignition (SI) and Direct Injection (DI). The database supports evaluations of health risks by categorizing toxicity based on specific assays and exposure conditions.

Compilation of database has revealed that several major gaps in the knowledge exists. Foremost, the toxicological studies cover almost only the fresh PM, and minority of studies investigated toxicity of aged PM including SecA (Künzi et al. 2015; Weitekamp et al. 2020; Delaval et al. 2025; Pardo et al. 2023). In addition, many toxicological publications lack the fundamental data for classification and integration of the results. This highlights the need for standardized reporting practices in studies examining transport sector emissions and exhaust toxicity. A recommended set of minimum parameters reporting parameters could include, amongst others: transport sector category, application type and details, exhaust aftertreatment, emission standards, test type (laboratory/RDE), test cycle, temperature, fuel category and details (sulfur and aromatic contents), CO_2, NO_x, NH_3, NMHC (preferably also aromatics), SO_2 and PM.

The toxicity rating system developed in PAREMPI provides a structured, semi-quantitative classification of biological responses to emissions. As for now, it was used to rate already existing toxicological studies in PAREMPI database. The four key endpoints included in the rating are:

- **C (Cytotoxicity):** Evaluates the reduction in cell viability following exposure to emissions. This indicates overall cellular damage and basic toxicity.
- **G (Genotoxicity):** Assesses the potential of emissions to cause DNA damage or mutations, which can contribute to long-term health risks such as cancer.
- **O (Oxidative Damage):** Measures oxidative stress responses, such as the generation of reactive oxygen species (ROS) or depletion of antioxidants like glutathione. These responses are linked to inflammation and chronic diseases.
- **D (Different / Other Endpoints):** Covers other relevant cellular effects not included above, such as immune response markers (e.g. cytokine release), changes in epithelial barrier integrity, or metabolic shifts.

Each endpoint is scored using the following standardized rating in Table 5. In final rating each emission condition is summarized using a combined rating code, e.g. C.1-G.0-O.x-D.nd, enabling quick cross-comparison of results across different sources or technologies. This system has been applied to published studies included in the PAREMPI database. Results from PAREMPI's own experimental toxicity assessments will be reported separately in Deliverables **D4.2** and **D4.3**.

Table 5. Standardized rating codes and their interpretations used in database

Code	*Interpretation*
nd	No data available
x	Unclear result
0	No harmful effect
1	Potentially harmful ($p < 0.05$)
2	Slightly harmful ($p < 0.01$)
3	Clearly harmful ($p < 0.001$)
4	Harmful with high confidence ($p < 0.0001$)
5	Very harmful with high confidence

5.1.4 Identification of gaps in knowledge and discussion

Despite significant progress in the PAREMPI project, several important knowledge gaps remain in the toxicological assessment of emissions—particularly in relation to secondary aerosols, newer technologies, and the integration of chemical and biological data.

Lack of integrated studies combining detailed chemical characterisation and toxicological testing, especially for SecA. Few studies have successfully combined in depth emission chemistry—including precursor gases, particle-phase composition, and atmospheric transformations—with wide toxicological endpoints such as cytotoxicity, genotoxicity, oxidative stress, and transcriptomics. This gap is particularly pronounced for secondary aerosols, which are increasingly recognized as critical contributors to health effects but remain poorly studied in controlled biological exposure setups. The challenge lies in the complexity of combining emission generation, photochemical aging (e.g. via OFRs), and direct biological testing within one workflow. Most projects are forced to focus on either emissions or toxicology due to differing technical demands, timelines, and expertise. Even when integration is attempted, not all components can be executed to full depth or resolution, limiting interpretability. This is especially true for under explored transport sectors such as aviation and marine, where toxicological data on SecA are virtually absent.

Underrepresentation of advanced biological models and endpoints: While advanced in vitro models and high-content endpoints like transcriptomic profiling provide more realistic and mechanistic insight into cellular responses, they remain underutilized in emission toxicity research. This is not only due to historical reliance on simpler monocultures and viability assays, but also an unfortunate consequence of the high logistical and financial complexity of tests. Such tests are expensive, time-consuming, and often inflexible—particularly in large-scale or field-based campaigns. For example, any malfunction during exposure (e.g. flow or power outage) can render cell material unusable for further analysis. This problem was encountered in the PAREMPI HD campaign, where results from a compromised exposure (Truck 2 with EN590) were still reported to reflect real-world conditions of testing on road.

Difficulty comparing results across studies due to methodological variability*:* Even where toxicological data are available, substantial heterogeneity in exposure systems, biological models, aerosol generation, and endpoint definitions complicates comparison across studies. Tools such as the PAREMPI toxicology (C-G-O-D) rating system can support cross-comparability, but wider adoption of standardised testing approaches is still needed.

Lack of toxicity data for modern technologies and alternative fuels: There is limited toxicological evidence for emissions from newer powertrains (e.g. hybrid, plug-in, electric-assisted) and low-emission or synthetic fuels (e.g. HVO, SPK blends). Furthermore, changes in fuel composition, engine technology, and after-treatment systems—and especially their evolving combinations—introduce complex interactions that can alter emission profiles in ways not yet captured by existing toxicological studies. These dynamic shifts in technology require significantly more research to assess not only individual components but also their integrated effects. Without well-designed studies reflecting real-world use cases, it remains unclear whether these innovations deliver meaningful reductions in health risk or simply meet emission standards on paper.

Across all campaigns, the biological component proved to be the most fragile part of the experimental setup. Advanced models like MucilAir™ and BEAS-2B offer valuable mechanistic insight but are highly sensitive to technical issues—once exposure begins, cells cannot be reused, making any malfunction a potential loss of data. SecA testing adds further complexity, as combining photochemical ageing with real-time ALI exposure limits the ability to run full endpoints across all samples. In some cases, compromises have to be made between exposure realism and biological depth. These challenges highlight the need for redundancy in biological workflows (e.g. backup

cultures, parallel runs) and for flexible planning when integrating emissions, chemical ageing, and high-content toxicology in a single campaign.

The toxicological impacts of secondary aerosols from traffic emissions are not well understood. There is a need for more systematic toxicological studies and standardized reporting practices to assess health risks and inform regulatory frameworks.

5.2 Health Impact Assessment and External Cost Evaluation of Air Pollution

5.2.1 Health Impact assessment

Health impact assessment (HIA) on mortality and morbidity attributed by population exposure to total secondary PM2.5 and related economical costs for externalities. Estimates under different scenarios supported by emerging toxicological and epidemiological studies.

Methods: HIA of air pollution quantifies the adverse health effects attributable to exposure in a given population, using established epidemiological relationships. The core of the methodology involves estimating the number of excess cases (e.g., premature deaths, hospital admissions) by integrating pollutant exposure levels, population data, baseline health statistics, and exposure–response functions (ERFs) derived from cohort or time-series studies.

The standard formula for HIA is:

$$\Delta Y = Y_0 \times (e^{\beta \times \Delta x} - 1) \times P$$

where:

ΔY = number of attributable cases (e.g., deaths), Y_0= baseline incidence rate of the health outcome, β = =concentration-response coefficient (ln(RR))/ Δx, Δx = change in pollutant concentration (e.g., µg/m³ of $PM_{2.5}$), P = exposed population, RR= relative risk associated with a unit change in pollutant concentration.

External cost evaluations then monetize these impacts using economic values such as:

- Value of a Statistical Life (VSL): commonly used for premature mortality,
- Value of a Life Year (VOLY): used in burden of disease and cost–benefit analyses.

These values are derived from stated preference studies or revealed preference methods and may be adjusted for local context or policy guidelines. Sensitivity analyses can be used to account for uncertainties in the ERFs, exposure estimates, population projections, and economic valuation.

Population exposure: To assess the populations exposure GIS (Geographical Information System) analyses were applied combining modelled annual levels of $PM_{2,5}$ for 2018 in combination with a population grid retrieved from Eurostat (GEOSTAT 1 km^2 population grid - Version 2018; 13 April 2021) using the software QGIS Desktop 3.42 and ArcMap 10.5.1. (Source: European Commission (Eurostat, Joint Research Centre and DG Regional Policy - REGIO-GIS.)

Exposure–Response Relationships for SecA: Within the PAREMPI project, we have explored whether conventional exposure–response functions for $PM_{2.5}$ are appropriate for use in health impact assessments involving SecA, or if current evidence justifies adopting component-specific risk estimates. This has been done in order to produce estimates under different scenarios.

While the association between fine particulate matter and mortality is well established, findings related to individual $PM_{2.5}$ components, such as secondary aerosols, remain inconsistent and uncertain, a point emphasized in several systematic reviews (Achilleos et al., 2017, Yang et al., 2019). The discussion below summarizes the most relevant literature and expert evaluations.

Scientific Evidence: Systematic reviews and recent cohort studies have provided mixed evidence on the health effects of individual $PM_{2.5}$ components, particularly secondary inorganic aerosols (SIA) such as nitrate, sulphate, and ammonium. For instance, Yang et al. (2019) reported a 2.6% increase in mortality per interquartile range (IQR) of nitrate (~2.3 μg/m³), while Karimi and Samadi (2023) estimated a 3% increase. However, SIA as a group was not significantly associated with mortality in the latter study. Cohort studies in North America and Europe have produced conflicting results. In California, long-term exposure to nitrate and sulphate was associated with elevated cardiovascular and all-cause mortality in a cohort of teachers, though these associations were not replicated in a subsequent study (Ostro et al., 2015). Canadian registry-based studies observed stronger associations with total $PM_{2.5}$ than with its individual components, although including component proportions improved model performance (Crouse et al., 2016). In the southeastern U.S., Wang et al. (2017) found lower relative risks for sulphate, nitrate, and organic carbon, suggesting that these components may be less harmful. Yet, a later satellite-based study from the same region found an increased mortality risk linked to nitrate, but not sulphate (Wang al., 2017). Similarly, Hao et al. (2023) reported that nitrate had a modest risk ratio (RR 1.023 per IQR increase), exceeding that of elemental carbon or organic matter, but falling below sulphate and ammonium (Hao et al., 2023). Findings from Nordic cohorts suggest a potential role of SIA in cardiovascular mortality, although the evidence for total mortality is less consistent (Hvidtfeldt et al., 2019, Nilsson Sommar et al., 2021). Despite growing interest, a major limitation in identifying component-specific effects lies in the availability and resolution of exposure data. Few studies have monitored or modelled specific $PM_{2.5}$ constituents across multiple sites, limiting the capacity to assess spatial variability, a key factor in long-term exposure assessments. Moreover, strong inter-correlations among components complicate the separation of individual health effects. Commonly analyzed constituents include secondary inorganic particles (nitrate, sulphate, ammonium), organic and elemental carbon, and trace metals. Exposure assessments frequently rely on land-use regression models or chemical transport models. While these methods offer improved spatial resolution, especially compared to earlier gridded models with resolutions from 1×1 km to 30 km, they introduce higher uncertainty and misclassification. Consequently, associations between individual components and health outcomes often lack precision or statistical significance, even when $PM_{2.5}$ overall is robustly linked to adverse effects.

Evaluations by Expert Bodies: Major public health agencies, including the World Health Organization (WHO), the U.S. Environmental Protection Agency (EPA), the UK's Committee on the Medical Effects of Air Pollutants (COMEAP), and France's ANSES—have consistently concluded that current evidence does not justify the use of component-specific exposure–response coefficients for $PM_{2.5}$. In their most recent evaluations, WHO's 2021 Air Quality Guidelines, the U.S. EPA's 2022 scientific review, and COMEAP's 2022 report all acknowledged potential toxicity differences among PM components but emphasized that the available evidence lacks the robustness required to support component-specific health effect estimates.

As a result, most health impact assessments treat all $PM_{2.5}$ mass equally, regardless of its chemical composition. Specifically, assessments typically do not differentiate between secondary aerosols (SecA) and other sources of $PM_{2.5}$. Nevertheless, some research indicates that the health risk associated with regional background $PM_{2.5}$ often dominated by secondary aerosols, may be lower per μg/m³ than that from local sources. For example, Turner et al. (2016) and Segersson et al. (2021) found that locally generated PM was more strongly associated with health outcomes than background PM.

In cases where exposure assessments specifically target SecA components, such as nitrate, using regionally derived risk estimates may be more appropriate. Yang et al. (2019) reported a 1.1% increase in mortality per µg/m³ of nitrate exposure, higher than the risk typically assigned to background $PM_{2.5}$ mass. However, this differentiation has rarely been implemented in practice in health impact assessments. To date, only one published health impact assessment has explicitly applied a separate risk estimate for nitrate (Hang et al., 2022). Similarly, the toxicity results from the PAREMPI project have not yet provided sufficiently robust or conclusive evidence to justify conducting a health impact assessment based on an alternative exposure scenario. While the findings offer important insights into potential mechanisms, the current level of uncertainty around the toxicological data limits their applicability for quantifying health effects in a policy-relevant context.

For this study, we therefore solely use the HR of 1.095 per 10 µg/m^3 increase, found in the update of the WHO AGQs from 2021 (Orellano et al. 2024).

In the HIA calculations in PAREMPI, we have employed country specific age-standardised mortality and baseline incidences for the whole of EU-27 for the following health outcomes, with relative risks recommended from the review done by Forastiere et al. (2024) for total SecA $PM_{2.5}$, see Table 6.

Table 6. Relative Risk Estimates for Disease Incidence from Long-Term Exposure to $PM_{2.5}$

Pollutant	Health Outcome (Incidence)	ICD-10 Codes	Age Group (Years)	Classification	Relative Risk (95% CI) per 10 µg/m³	Exposure Range (µg/m³)
$PM_{2.5}$	Asthma in children	J45	0–18	A	1.34 (1.10–1.63)	5–38
	Chronic Obstructive Pulmonary Disease (COPD)	J41–J44	30+	A	1.18 (1.13–1.23)	5–26
	Ischemic Heart Disease (IHD) Events	I21–I22	30+	A	1.13 (1.05–1.22)	5–65
	Stroke	I60–I64	30+	A	1.16 (1.12–1.20)	5–36
	Type 2 Diabetes	E11–E14	30+	B+	1.10 (1.03–1.18)	5–79
	Dementia	F00–F03, G30	60+	B+	1.46 (1.20–1.78)	5–25
	Autism Spectrum Disorders (ASD)	F84.0–F84.9	2–12	B+	1.66 (1.23–2.25)	5–30
	Lung Cancer	C34	30+	A	1.16 (1.10–1.23)	5–44

Notes:

- **Classification**: 'A' indicates a reliable quantification of health effects is possible in an HRA; 'B+' denotes that HRA is possible, but there is greater uncertainty around the reliability of the CRF compared to those included in list A.
- **Exposure Range**: Represents the range of mean exposures in the studies from which the relative risks were derived. Applying these CRFs outside these ranges may introduce additional uncertainty.
- **Systematic Review References**: Detailed references can be found in the original article.

The total annual number of deaths and morbidity outcomes attributable to SecA across the EU-27, as well as per country, are presented in Table 7. According to estimates, approximately **111,000 premature deaths** per year in the EU-27 can be attributed to SecA exposure. In addition, SecA is associated with an estimated:

- **22,940** autism spectrum disorder (ASD) cases
- **206,301** acute myocardial infarctions
- **43,681** lung cancer cases
- **104,049** stroke cases
- **100,921** type 2 diabetes cases
- **591,001** dementia cases
- **203,945** chronic obstructive pulmonary disease (COPD) cases
- **185,818** childhood asthma cases

Table 7. Estimated population weighted exposure to total secondary aerosols from various sources, including traffic, across the EU-27 and attributed mortality and morbidity. Abbreviations: ASD – Autism Spectrum Disorders; AMI – Acute Myocardial Infarction; COPD – Chronic Obstructive Pulmonary Disease. For dementia, we have used the population above 65 years of age, for childhood asthma, the population 15 years or younger. For autism the whole population. For COPD, stroke, diabetes, AMI, lung cancer and Acute Myocardial Infarction, assuming 70% of the population is 30 years or older across the EU.

Country	SecA (µg/m³)	Deaths	ASD	AMI	Lung cancer	Stroke	Diabetes	Demens	COPD	Asthma child
Austria	10,4	2647	581	4745	927	2743	1616	7285	6189	3862
Belgium	10	3455	810,2	5173	1528	2802	2921	9736	7503	5511
Bulgaria	6	2341	216	3944	355	2655	1469	2794	2097	1983
Croatia	7,5	1099	158	1792	405	1087	1047	2048	1716	1474
Cyprus	6,1	154	39	130	52	76	206	287	267	282
Czechia	11,2	4021	924	10268	1130	4018	4756	8312	6653	6973
Denmark	6,8	1103	276	1177	590	867	883	2063	2686	1973
Estonia	4,3	199	40	785	61	165	155	428	222	280
Finland	3,3	568	98	1325	190	699	624	1531	1107	900
France	6,4	11917	2827	27404	5830	10133	8631	289994	24558	24326
Germany	11,1	26406	5554	56326	10735	28224	23033	101180	64498	32950
Greece	5,8	2187	364	2789	940	2782	1628	7113	5002	2432
Hungary	8,5	3374	531	8064	1377	2824	3073	5474	5248	4059
Ireland	3,3	477	147	660	200	255	387	801	938	1192
Italy	9,3	16476	3783	28282	7232	14082	16585	82178	42421	23003
Latvia	4,9	427	58	983	85	507	245	662	370	484
Lithuania	5,7	646	92	1744	137	840	318	1027	620	669
Luxembourg	9,3	141	34	129	51	78	122	203	270	279
Malta	4,7	67	15	79	20	45	87	153	620	102
The Netherlands	10,7	5284	1305	8028	2791	4372	4315	14264	270	6154
Poland	7,7	11015	1872	14594	3821	8155	10801	17239	13260	39575
Portugal	4,6	1308	216	968	428	1146	1780	4109	2834	2128
Romania	6,4	5327	719	10169	1170	6028	2922	7252	6056	8057
Slovakia	9	1954	351	3660	388	1708	1403	2358	1982	2595
Slovenia	9,4	599	129	1339	258	469	577	1413	1011	1403
Spain	5,2	6641	1416	9096	2586	5876	9973	16661	1639	9222
Sweden	4,6	1420	385	2648	394	1413	1364	4436	3908	3950
EU-27		**111257**	**22940**	**206301**	**43681**	**104049**	**100921**	**591001**	**203945**	**185818**

5.2.2 Description of external costs evaluation based on health impact assessment

In the PAREMPI project, we adopted the European Commission's standard Value of a Statistical Life (VSL) of €4,57 million for estimating mortality-related external costs, reflecting a well-established benchmark used in EU policy assessments and ensuring consistency with cost-benefit analyses in the environmental and public health sectors. We furthermore estimated external costs per case for various health outcomes below see the ones relevant to health impact assessments (HIAs) within the European Union (EU). These estimates encompass both direct costs (such as healthcare and social services) and indirect costs (including productivity losses and informal care). It's important to note that these figures are approximate and can vary based on country, healthcare system, disease severity, and methodological approaches. Table 8 represents the estimated external costs per case in EU used in PAREMPI.

Table 8. Estimated External Costs per Case in the EU

Health Outcome	*Estimated Cost per Case (€)*
Asthma in Children	*€1,000*
Chronic Obstructive Pulmonary Disease (COPD)	*€6,000*
Ischemic Heart Disease (IHD)	*€10,000*
Lung cancer	*36,000*
Stroke	*€25,000*
Type 2 Diabetes	*€7,500*
Dementia	*€40,000*
Autism Spectrum Disorder (ASD)	*€17,000*

Notes:

- Asthma in Children: Costs are influenced by the frequency of exacerbations and the level of asthma control.
- COPD: Costs escalate with disease severity, with severe cases incurring higher hospitalization and medication expenses.
- IHD and Stroke: These cardiovascular conditions impose significant economic burdens due to treatment costs and loss of productivity.
- Type 2 Diabetes: Management costs include ongoing medical care and potential complications leading to increased healthcare utilization.
- Dementia: The majority of costs are attributed to long-term care needs and informal caregiving.
- ASD: Early and intensive behavioral interventions can be costly but may lead to long-term savings by improving outcomes.

The total annual societal cost of premature mortality attributable to SecA in the EU-27 is estimated at approximately **€508.5 billion**, based on PAREMPI calculations (Table 9) using a value of a statistical life (VSL) approach. When the monetized costs of selected morbidity outcomes are also included, the total increases by **€35.2 billion** to approximately **€544 billion**.

It should, however, be noted that estimates of morbidity-related costs vary substantially across studies because they depend strongly on the choice of health endpoints, valuation method, time horizon, and cost perspective (for example, healthcare costs only versus broader societal costs including productivity losses and informal care). Recent work in the VALESOR project within the METEOR cluster suggests higher morbidity cost estimates than those applied here, indicating that the PAREMPI estimates should be regarded as **conservative**.

At the same time, the combined total of mortality and morbidity costs should be interpreted with caution. Current methodological guidance distinguishes clearly between mortality valuation using **VSL/VOLY** and morbidity valuation using **cost-of-illness** or willingness-to-pay approaches. OECD guidance notes that mortality welfare costs valued with VSL exclude morbidity impacts, while cost-of-illness estimates may include direct medical costs as well as indirect costs such as productivity losses. This creates a risk of **partial overlap**, particularly if morbidity cost estimates include components related to fatal cases or to productivity losses that are also implicitly captured in mortality valuation. In addition, disease endpoints such as COPD, stroke, myocardial infarction and dementia are not independent, and individuals may experience multiple conditions over time. For these reasons, mortality and morbidity costs are often best presented **separately**, or, if summed, described as an indicative aggregate that may involve some degree of double counting.

Table 9. Estimated societal costs in Million Euros due to total secondary aerosols from various sources, including traffic, across the EU-27. Abbreviations: ASD – Autism Spectrum Disorders; AMI – Acute Myocardial Infarction; COPD – Chronic Obstructive Pulmonary Disease.

Country	Deaths	ASD	AMI	Lung cancer	Stroke	Diabetes	Dementia	COPD	Asthma child
Austria	7940,1	64,277	47,45	9,877	68,575	12,12	291,4	37,134	3,862
Belgium	10365,6	83,912	51,73	13,7734	70,05	21,9075	389,44	45,018	5,511
Bulgaria	7022,4	56,848	39,44	3,672	66,375	11,0175	111,76	12,582	1,983
Croatia	3297	26,69	17,92	2,686	27,175	7,8525	81,92	10,296	1,474
Cyprus	462	3,74	1,3	0,663	1,9	1,545	11,48	1,602	0,282
Czechia	12064,5	97,665	102,68	15,708	100,45	35,67	332,48	39,918	6,973
Denmark	3311,7	26,809	11,77	4,692	21,675	6,6225	82,52	16,116	1,973
Estonia	598,5	4,845	7,85	0,68	4,125	1,1625	17,12	1,332	0,28
Finland	1705,2	13,804	13,25	1,666	17,475	4,68	61,24	6,642	0,9
France (metropolitan)	35750,4	289,408	274,04	48,059	253,325	64,7325	11599,76	147,348	24,326
Germany	79218,3	641,291	563,26	94,418	705,6	172,7475	4047,2	386,988	32,95
Greece	6560,4	53,108	27,89	6,188	69,55	12,21	284,52	30,012	2,432
Hungary	10122	81,94	80,64	9,027	70,6	23,0475	218,96	31,488	4,059
Ireland	1432,2	11,594	6,6	2,499	6,375	2,9025	32,04	5,628	1,192
Italy	49427,7	400,129	282,82	64,311	352,05	124,3875	3287,12	254,526	23,003
Latvia	1281	10,37	9,83	0,986	12,675	1,8375	26,48	2,22	0,484
Lithuania	1938,3	15,691	17,44	1,564	21	2,385	41,08	3,72	0,669
Luxembourg	422,1	3,417	1,29	0,578	1,95	0,915	8,12	1,62	0,279
Malta	201,6	1,632	0,79	0,255	1,125	0,6525	6,12	3,72	0,102
The Netherlands	15852,9	128,333	80,28	22,185	109,3	32,3625	570,56	1,62	6,154
Poland	33045,6	267,512	145,94	31,824	203,875	81,0075	689,56	79,56	39,575
Portugal	3924,9	31,773	9,68	3,672	28,65	13,35	164,36	17,004	2,128
Romania	15981	129,37	101,69	12,223	150,7	21,915	290,08	36,336	8,057
Slovakia	5863,2	47,464	36,6	5,967	42,7	10,5225	94,32	11,892	2,595
Slovenia	1797,6	14,552	13,39	2,193	11,725	4,3275	56,52	6,066	1,403
Spain	19922,7	161,279	90,96	24,072	146,9	74,7975	666,44	9,834	9,222
Sweden	4260,9	34,493	26,48	6,545	35,325	10,23	177,44	23,448	3,95
EU-27	**333769,8**	**2701,946**	**2063,01**	**389,9834**	**2601,225**	**756,9075**	**23640,04**	**1223,67**	**185,818**

5.2.3 Identification of gaps in knowledge and discussion

Several gaps in the knowledge regarding health impact assessment were identified and listed below.

Uncertainty in SecA modeling: Most likely that the models still underestimate the SOA formation from many anthropogenic VOCs since only part of known oxidation pathways and oxidation products are included in the models. Also oxidation products volatility distributions are still highly uncertain especially at high NO_x-conditions and cold temperatures.

Uncertain Toxicity of SecA: It remains unclear whether the health effects of SecA are comparable to those of other $PM_{2.5}$ components, such as black carbon (BC). While several studies have examined associations between SecA and including components and mortality (with large heterogeneity in results), there is a significant lack of studies regarding morbidity outcomes, including cardiovascular and respiratory diseases.

Assumptions in Health Impact Assessments: Due to limited component-specific evidence, current health impact assessments typically assume that the toxicity of SecA is equivalent to that of total $PM_{2.5}$, such as here. This may lead to either overestimation or underestimation of health risks, depending on the true toxicity profile of SecA.

Limited Demographic Data: A lack of detailed population statistics—such as age-specific population distributions (e.g., population aged 30 years and older) for EU-27, hampers efforts to accurately estimate disease burden and exposure impacts across subgroups.

Unclear Source Contributions: The relative toxicity of different sources (e.g., traffic, agriculture, industry) remains poorly characterized. This limits the ability to develop targeted mitigation strategies and to assess the health implications of source-specific exposures.

Influence of Meteorological and Regional Factors: The considerable heterogeneity observed in the literature may be explained by variations in aerosol composition, meteorological conditions (e.g., temperature, humidity, wind patterns), and population susceptibility. These factors are not consistently accounted for in existing studies, making it difficult to generalize findings across regions.

Gaps in the knowledge regarding external costs evaluation were identified and listed below.

Country-specific estimated costs: A key limitation in our analysis is the use of EU-wide average values for the Value of a Statistical Life (VSL) and estimated costs per case. While this approach ensures comparability across countries and simplifies the estimation process, it introduces uncertainty due to the substantial variation in economic conditions, healthcare costs, and societal preferences across EU member states. Country-specific valuations would more accurately reflect national contexts and improve the precision of health impact cost estimates. However, due to data availability and consistency issues, we opted for harmonized EU-level estimates. This choice enables cross-country comparison but may under- or overestimate the true economic burden in individual countries.

According to new estimates done in PAREMPI project the estimated the societal costs of SecA across the EU amount to approximately €544 billion annually. Although these estimates carry inherent uncertainties—due to variations in exposure, health outcomes, and cost assumptions—they underscore the urgency of identifying and implementing cost-effective mitigation strategies.

PART III: Prevention of SecA: Prevention technologies of the harmful precursor emissions to mitigate SecA formation in urban areas

6 Potential SecA prevention technologies

6.1 Introduction to prevention technologies

Reducing exhaust emissions, especially those that contribute to secondary aerosol (SecA) formation, requires a multifaceted approach integrating, amongst other, policies, legislation and technical innovations. This section focuses on technological measures, i.e. ***exhaust after-treatment (EAT) systems****,* ***fuel innovations, and engine design improvements,*** *which can significantly lower emissions of SecA precursor gases, i.e., certain VOCs, NO_x, NH_3, and SO_x, as well as directly emitted PM. This section draws conclusions from PAREMPI results and supporting literature. PAREMPI D5.4 "Transport policy recommendations" will translate these findings into the policy recommendations and discuss prevention measures further in a broader perspective.*

Secondary aerosols are formed through atmospheric reactions involving precursor gas emissions from transport sources, VOCs, NO_x, NH_3 and SO_x, leading to organics, nitrates, ammonium and sulfates through subsequent atmospheric reactions. **From VOCs**, SecA formation is attributed especially to monoaromatic compounds, primarily originating from unburned and partially burned fuel and lubricating oil (see Sections 1.2 and 1.3). **NO_x emissions** are formed at high combustion temperature from intake air nitrogen and oxygen, especially in diesel engines using the excess air in combustion, but also in stoichiometric spark-ignited (SI) engines. **NH_3 emissions** from transport applications are formed in some EAT systems, and in future applications potentially by ammonia-fueled engines. **Sulfur**, present in fuel and lubricating oil, is primarily combusted to **SO_2**. A few percents of SO_2 is oxidized to SO_3, which further reacts with water vapor to form sulfuric acid and exhaust sulfate particles. In addition, oxidative exhaust after-treatment systems can increase the conversion of SO_2 to SO_3 and thus sulfuric acid, which can affect particulate sulfate concentrations and e.g. nanoparticle formation in cooling exhaust and thus nanoparticle concentrations in traffic-influenced areas.

Exhaust after-treatment technologies are particularly effective in controlling gaseous SecA precursors. For gasoline spark-ignition engines operating at stoichiometric air-fuel ratios, three-way catalysts (TWCs) are the standard, simultaneously reducing NO_x, CO and organic compounds in the exhaust, but they can also induce NH_3 formation. In the diesel engines, selective catalytic reduction (SCR) and lean NO_x traps (LNTs) are commonly used to reduce NO_x emissions. The SCR systems use urea-derived ammonia to convert NO_x into harmless nitrogen and water. **NH_3 emissions** from transport applications can be formed in the EAT systems, such as urea-based SCR potentially slipping NH_3 depending on system control, and especially from the TWC aftertreatment (Mejía-Centeno et al. 2005, Suarez-Bertoa et al. 2016). To prevent excess ammonia from escaping into the atmosphere, an ammonia slip catalyst (ASC) is often installed downstream the SCR. In contrast, TWCs are generally not combined with NH_3 mitigation measures.

Other EATS components include the diesel oxidation catalyst (DOC) reducing CO, hydrocarbons, and the organic fraction of PM. The diesel particulate filters (DPFs) and the gasoline particulate filters (GPFs) reduce particle number emissions efficiently.

Fuels have a role in forming SecA even for low-emission technologies, especially during the warm-up phase of both the engine and exhaust after-treatment systems. Fuel standards addressing monoaromatic contents of fuels would support air quality goals. Additionally, fuels having high hydrogen to carbon ratio, e.g. methane and paraffinic fuels, burn cleanly and lower combustion temperatures, which in turn reduces NO_x formation.

Engine internal technologies that reduce the amount of unburned fuel can assist lowering the potential for SecA formation. Modest reductions in engine-out NO_x emissions can be achieved through engine design strategies, such as optimization of injection system, turbochargers, the charge air cooling and variable valve timing. A widely used strategy is exhaust gas recirculation (EGR), which redirects a portion of exhaust gases back into the intake air. This lowers combustion temperatures and suppresses NO_x formation. However, EGR can negatively impact fuel economy (Kim et al., 2019).

6.2 Findings on SecA and PM_{aged} emissions prevention technologies

Light duty cars

The PAREMPI results highlight that tightening emission standards have cleaned the car emissions (sections 3.1 and 4.2). For cars, Simon et al. (2025) revealed that while new cars emit low overall particle emissions, cold-start conditions remained to trigger SecA formation. This aligns with Simonen et al. (2019), who found organic compounds dominating SecA after cold-start, and by ammonium, sulfate, and nitrate as the engine warmed. Despite of being modern cars, Euro 6dT CNG car emitted unexpectedly high PM_{aged} emissions attributed to its gasoline backup system, and Euro 6d PHEVs emitted elevated particle levels when switching from electric to combustion mode under dynamic driving.

EATS. The role of particle filtration was evident in PAREMPI results. PM_{fresh} emissions were high for gasoline cars lacking GPFs, especially in cold conditions, while cars with filtration emitted very low particle emissions, though regeneration events may occasionally lead to elevated emissions. Diesel cars are equipped with SCR, LNT or other NO_x reduction technologies. TWCs are effective at reducing NO_x and NMOG emissions, while they can emit NH_3 enhancing SecA formation (e.g. Euro 6b and Euro 6d PHEV gasoline, and CNG cars) (Simon et al., 2025). The Euro 6b gasoline car promoted ammonium nitrate (NH_4NO_3) formation, while the Euro 4 diesel car promoted ammonium sulfate ($(NH_4)_2SO_4$) formation. In NH_3-limited urban environments, gasoline vehicles may act as NH_3 sources, enhancing SecA formation, as addressed by Simon et al. (2025). NH_3 is currently unregulated for cars, and future standards would benefit from stringent and harmonized NH_3 limits.

Fuel composition influence emissions and SecA formation, particularly through their monoaromatic content and sulfur contents. Gasoline typically contains 1-ring aromatics contributing to SecA formation. Saarikoski et al. (2024) demonstrated that aromatic-free fuels significantly cut down non-volatile, fresh, and aged particles. In Europe, fuel standards limit aromatic content of gasoline to a max. 35.0 vol% and benzene to max. 1.0 vol% (EN 228:2012 + A1:2017). For diesel fuel (EN590:2022), there is no limit on total aromatics, though polycyclic aromatic hydrocarbons (PAHs, di+) are limited to max. 8.0 wt%. Diesel fuels typically contain heavier aromatics than gasoline; hence the amount of monoaromatic SecA precursors may be low. Fuel standards could be much tighter as regards aromatic contents of gasoline and diesel fuel in Europe.

Low-sulfur fuels effectively reduce SO_x emissions. Sulfur levels in road fuels in Europe are already minimal (max. 10 mg/kg), which is a significant achievement in reducing sulfur-related pollution. The sulfur content in lubricating oils may still contribute to residual SO_2 emissions, a factor worth of further investigation, especially in applications where oil consumption is significant.

Fuels such as methane, paraffinic fuels like hydrotreated vegetable oil (HVO) and gas-to-liquid (GTL) fuels, as well as alcohols and ethers, have high hydrogen to carbon (H/C) ratios, burn cleanly and lower combustion temperatures, which in turn reduces NO_x formation (Kuronen et al., 2007; Murtonen et al., 2009; Ushakov et al., 2019). However, not all biofuels offer the same benefits. For example, fatty acid methyl esters (FAME) biodiesels have shown a slight increase in NO_x emissions compared to conventional diesel (Yusuf et al., 2022).

Engine internal technologies that reduce the amount of unburned fuel can help lower the potential for SecA formation. Modest reductions in engine-out NO_x emissions can be achieved through engine design, e.g. optimization of injection, turbochargers, charge air cooling, variable valve timing and EGR (Kim et al. 2019).

Heavy-duty vehicles

For heavy-duty applications, trends mentioned for light-duty cars apply as regards the effect of emission standards, fuel composition, EATS and engine internal technologies. In the PAREMPI study, two Euro VI heavy-duty trucks (older step A and a newer step E), both equipped with modern exhaust EAT systems, demonstrated exceptionally low PM_{aged} emissions, especially the Euro VI E truck. The results confirm efficiency of stringent emission standards in reducing PM_{aged} emissions from HD engines. While NO_x, NMOG and NH_3 emissions were below emission limits for the studied vehicles, cold start emissions were elevated.

PAREMPI did not assess engine-out emissions, but existing literature provides references. For instance, Karjalainen et al. (2019) reported engine-out PM_{aged} emissions of the heavy-duty diesel engine as high as 106.2 mg/kWh and PM_{fresh} at 44.7 mg/kWh. After EATS, these values dropped dramatically to below 1 mg/kWh. PM_{fresh} was primarily composed of black carbon, while PM_{aged} contained more sulfates and organic compounds. Interestingly, fuel type had only a minor influence on SecA formation, suggesting that lubricants or combustion processes themselves may be key sources of precursors. Supporting this, Gordon et al. (2014) found that Class 8 HD trucks running on fuel with 12% aromatics produced high PM_{fresh} emissions; however, when equipped with diesel oxidation catalysts and DPF, emissions dropped to below 2 mg/kWh. These findings underscore the impact of EATS, which can reduce both fresh and aged PM emissions by orders of magnitude, significantly mitigating the adverse environmental impacts of heavy-duty vehicles.

Table 10 represents a summary of technologies to prevent SecA formation and PM_{aged} emissions from road transport sector.

Table 10. Summary of technologies to prevent SecA formation and PM_{aged} emissions from road transport.

EATS technologies	Fuel technologies	Engine internal
NMOG: EAT with oxidation function, such as TWCs and DPFs, reduce NMOG. EAT may not perform optimally during cold-starts and PHEVs switching to ICE mode.	**Fuel standards:** Tight limits for fuel (mono)aromatic content would reduce SecA formation, especially during engine and EAT warm-up. In Europe, high content of aromatics is allowed in gasoline and diesel fuels.	Engine internal technologies that reduce the amount of unburned fuel can help lower the potential for SecA formation.
NOx: TWC is typical in SI cars, while SCR and LNT in diesel cars. EAT may not perform optimally during cold-starts and PHEVs switching to ICE mode.	Fuels like methane, paraffins, alcohols and ethers reduce engine-out NO_x by lowering combustion temperature.	Modest reductions in engine-out NO_x through engine design, e.g. injection, turbochargers, charge air cooling, variable valve timing and EGR.
NH_3: ASC can be installed to remove excess NH_3 from SCR. ASC not typical with TWCs.	–	–
SO_2: EAT systems are not applied for SO_2 reduction from cars and vehicles.	In Europe, minimal sulfur contents of road fuels, max. 10 ppm, reduce SO_x. Low-sulfur lubricating oils could assist this aim.	–
PM_{fresh}: emissions are primarily reduced by particle filters (DPF, GPF).	Fuels having low aromatic content, high H/C-ratio and oxygen can reduce PM_{fresh} emissions.	–

Marine sector

The PAREMPI simulations using a diesel generator and marine fuels revealed substantial particulate matter emissions (fresh and aged), alongside fluctuating levels of precursor gases.

Emission standards. The International Maritime Organization (IMO) regulates NO_x emissions, limits depending on engine size and type. Compliance with IMO Tier III NO_x limits in NO_x Emission Control Areas (NECAs) for newbuilds is typically achieved with SCR and dual-fuel LNG engines (LNG DF). Modest reductions are achieved with technologies such as methanol combustion, EGR, and SO_x scrubbers. Tight emission standards are applied in inland waterways.

Fuel composition. Marine fuels vary by grades as defined by the ISO 8217 standard. Sulfur contents in marine fuels have historically been high; under current regulations, the global sulfur limit is 0.5 wt%, while a stricter limit of 0.1 wt% applies within Sulfur Emission Control Areas (SECAs). SO_2 emissions reflect the sulfur content of fuels. Elevated fuel sulfur levels shift composition of PM_{aged} toward sulfates, indicating sulfur-driven SecA formation, as also observed in PAREMPI.

Conventional marine fuels generally contain heavier fractions than road transport fuels. In PAREMPI, the use of distillate marine fuel (DMB grade) resulted in elevated NMOG emissions, and organic material was observed in both fresh and aged PM, potentially originating from unburned or partially burned heavy organic fraction of fuel. Conventional marine fuels typically contain substantial amounts of aromatics, and may contain even asphaltenes; for example, a marine distillate fuel had 42.6 wt% aromatics (Aakko-Saksa et al. 2016), although monoaromatics may be absent. Notably, heavy aromatic compounds are of concern due to their potential contribution to increased exhaust toxicity.

Clean marine fuels reduce emissions efficiently, especially methane in LNG DF engine concept and methanol, have shown to substantially reduce NMOG and NO_x emissions. Fuels enabling low NO_x emissions are important, since EAT systems are not consistently applied with marine engines, and not commonly used outside of emission control areas. LNG LPDF and SI engines can meet Tier III

NOx limits without SCR, unlike e.g. LNG HPDF (Pavlenko et al. 2020). Methanol DF engines emit less NO_x than diesel engines without SCR, but more than LNG DF.

EAT. Exhaust aftertreatment technologies are available for marine engines to mitigate NO_x, NH_3, SO_x and PM_{fresh} emissions, and some of these reduce also NMOG emissions. SCR can achieve NO_x reduction efficiencies exceeding 90% and is compatible even with low-quality marine fuels (Di Natale and Carotenuto 2015, Jayaram et al. 2011). The marine sector is the only transport mode that applies EAT specifically for SO_x removal. Scrubbers achieve up to 99% SO_x reduction, outperforming even 0.1 wt% sulfur fuels (Winnes et al. 2010), and enable continued use of high-sulfur fuels. SO_x scrubbers can slightly reduce NO_x by dissolving NO_2 into the scrubber water; however, this effect is limited to max. 12% to prevent nitrate discharge into the marine environment (MEPC.259(68)). Ammonia slip catalyst downstream SCR remove excess NH_3, while scrubbers may enable slight removal through dissolving some NH_3 in the scrubber water. NMOG emissions can be mitigated by marine DOCs and DPFs, and also by SCR to some extent. In practice, SCR and scrubber technologies are commonly used in emission control areas, while DOC and DPF remain limited despite their availability.

In PAREMPI, EAT was not applied in the test setup mimicking marine exhaust. However, NH_3 injection into the exhaust was used to mimic NH_3 slip from SCR, or unburned NH_3 from ammonia-fueled engine. The presence of NH_3 (60 ppm) in exhaust led to remarkable PM_{aged} emission dominated by nitrates, and significant contributions of ammonium and organic components. In the absence of NH_3 injection, NH_3 levels remained near the detection limit. Consistent with these findings, previous studies report low NH_3 emissions downstream of SCR, indicating effective ammonia slip control (Aakko-Saksa et al., 2023). However, the introduction of ammonia as marine fuel highlights the need for future regulatory limits on NH_3 emission.

Only a few of studies have explored PM_{aged} emissions from ships. An example is reported by Karjalainen et al. (2022), who performed onboard measurements on a Roll-on/Roll-off Passenger ship equipped with a DOC and a scrubber operating within a Sulfur Emission Control Area (SECA). Their findings showed negligible PM_{aged} emissions, suggesting that either the DOC, the scrubber, or their combination was effective in eliminating particulates and SecA precursors. The scrubber alone was shown to reduce SO_x concentrations by over 99%. The same study also revealed that engine-out PM emissions were higher when the ship operated on residual fuel compared to distillate fuel. During open-sea cruising, PM emissions reached 1.7 g/kg of fuel, with composition primarily of sulfates (66%), followed by organic compounds (30%) and a small fraction of black carbon (4%). These results indicate the potential benefits of transitioning to cleaner marine fuels.

Engine internal technologies that reduce the amount of unburned fuel can mitigate the potential SecA formation. However, reductions in engine-out NO_x emissions are typically modest, e.g. optimization of injection, turbochargers, charge air cooling, variable valve timing and EGR (Kim et al. 2019), if not combined with clean fuels.

Table 11 represents a summary of potential of different technologies to prevent SecA precursor emissions from the marine sector.

Table 11. Potential of different technologies to prevent precursor emissions for SecA formation in marine sector.

EATS	Fuels	Other
NMOG: Marine DOCs and DPFs reduce NMOG emissions, and SCR possesses modest oxidative function. DOC and DPF are available for shipping, but they are not commonly used.	Marine fuels vary by grades (ISO 8217). Marine fuels contain aromatics; however, likely heavier than monoaromatics. Aromatic content potentially affect exhaust toxicity.	Engine internal technologies that reduce the amount of unburned fuel can lower the potential for SecA formation.
NO_x: SCR can reduce NO_x emissions by over 90%. SO_x scrubbers may slightly reduce NO_x by dissolving NO_2 into the scrubber water (max. 12%).	Fuels enabling low NO_x emissions include LNG and methanol. LPDF and SI engines can meet Tier III NO_x limits without SCR, unlike e.g. LNG HPDF.	Engine-out NO_x can be modestly reduced through engine design.
NH_3: ASC downstream SCR can remove excess NH_3. SO_x scrubbers may reduce NH_3 slightly (dissolves in scrubber water).	Future ammonia engines may emit NH_3 emissions, requiring dedicated mitigation strategies.	–
SO_2: Scrubbers enable up to 99% SO_x reduction, outperforming even 0.1 wt% sulfur fuels.	Reducing sulfur contents of marine fuels is efficient technology to reduce SO_x emissions. Currently marine fuels containing sulfur up to 0.5 wt% globally, and up to 0.1 wt% in SECA.	–

Aviation sector

In aviation, fuel composition plays a central role in determining exhaust emissions, including those that contribute to SecA formation. This is largely because aircraft engines operate without EAT systems due to operational and design constrains, making fuel composition an important factor in mitigating emissions.

Aviation fuel standards allow high aromatic contents; ASTM D1655 for Jet A-1 fuel sets aromatic limit of max. 26.5 vol%, and ASTM D7566 with synthesized hydrocarbons requires min. 8 vol% aromatics. Aviation sector permits fuels with high sulfur contents of up to 0.3 wt%, and SO_2 levels are expected to reflect these sulfur levels. Tighter fuel standards could enable emission reductions in aviation sector, provided that safety and material aspects are not compromised. Paraffinic SAF fuels could enable improving jet fuel properties to achieve low emissions. For example, paraffinic fuels can moderately reduce NO_x emissions and they typically do not contain sulfur. In addition, turbine design strategies could reduce NO_x emissions, and the amount of unburned fuel, thereby mitigating the SecA formation.

In PAREMPI, using a CAST burner and aviation fuels mimicking idle operating conditionsshowed high PM_{aged} emissions, dominated by the organic fraction, along with elevated NMOG emissions. These findings emphasise the need to improve emission reduction strategies in aviation to improve air quality near airports.

Table 12 represents potential of different technologies to prevent SecA precursor emissions from the aviation sector.

Table 12. Potential of different technologies to prevent precursor emissions for SecA formation in aviation sector.

	Fuels	Other
NMOG	Aromatic limits are currently high for jet fuels.	Engine technologies reducing the amount of unburned fuel can lower the SecA formation.
NO_x	Fuels capable to reduce NO_x include for example paraffinic fuels.	Modest reductions in NO_x emissions could be achieved through design strategies.
NH_3	–	–
SO_2	Aviation sector permits fuels with high sulfur contents up to 0.3 wt%.	–

6.3 Robust measurement system to generate data to support policies

Robust and reliable measurement methods facilitate the generation of a comprehensive knowledge base on aged particulate matter, and its precursor emissions, covering a range of transport technologies. This information is necessary for supporting policies aimed at limiting and preventing ambient PM levels originating from transport sources. The results are also needed for the modelling activities and health impact assessments. The term "robust measurement system" refers to its applicability in real-life field conditions, such as on-road and on-board environments, without the need for dedicated specialists, although expertise in emission measurements is still required. Measuring the PM_{aged} formed through secondary reactions of precursor gases in the atmosphere remains challenging with current scientific methods. These techniques are often unsuitable for on-road or on-board applications due to their large size, complexity, and sensitivity to harsh conditions. However, smaller instruments are being developed, such as PEMS-type FTIRs, portable oxidative flow reactors (OFR), some of which are already available. To note, SecA is not directly measured, while it is calculated difference between measurable "tailpipe" emission PM_{fresh}, and PM_{aged}, the total potential particle mass formed through atmospheric reactions.

The robust measurement system for quantifying potential PM_{aged} emissions and gaseous precursors forming secondary aerosols is suggested in PAREMPI **D2.3** report. This system comprises of a portable Oxidation Flow Reactor (OFR), a dilution system, a particle number size distribution (PSD) device, from which the results are converted to PM mass (PN to PM mass), instruments for measuring precursor gases (NO_x, NH_3, SO_x, aromatics) that potentially form SecA as well as other necessary gases (NO_x, CO, CO_2) required to define settings for the OFR and to calculate emission factors and toxicity measurement (Figure 19, remarks in Appendix 3). The recommended emission metric for PM_{aged} is mass per megajoule, defined based on the CO_2 concentrations and fuel properties, eliminating the need for exhaust or fuel flow measurements and therefore simplifying the setup. The robust measurement methods were evaluated alongside a complex measurement system within the PAREMPI project. Although not detailed here, regulated PM is also an important parameter, enabling the presentation of the ratio of aged to fresh PM.

The same robust setup could be used regardless of the transport sector studied, although different dilution ratios are needed, and instruments may vary in the setups. The outlook beyond PAREMPI encompasses the harmonisation of the methods and protocol, precise definition of conditions and the target settings for the OFR, as well as the detailed requirements for instruments. These definitions are critical to ensure comparability of results across different studies.

Toxicity measurements in real-world conditions are essential to gain holistic view on the impact of exhaust on health. In the PAREMPI, the integration of a portable-size Air-Liquid Interface (ALI) exposure chamber into a moving vehicle, enabling on-road sampling of heavy-duty truck emissions,

and in situ toxicity testing, was a significant achievement. In the PAREMPI **D2.3** report, robust measurement system is suggested, along with the robust toxicity setup. In this report, toxicological assessment of transport-derived exhaust with the robust measurement system suitable for real-life conditions is described in section 5.1.1. and 5.1.2.

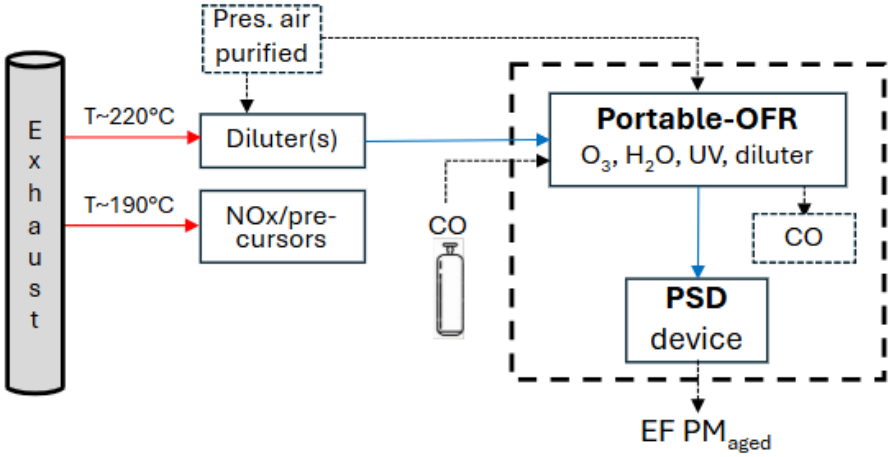


Figure 19. Schematic layout of the robust setup suggested for measuring PM_{aged}.

6.4 Identification of gaps in knowledge and discussion

Link between precursor emissions and PM_{aged} composition was evident in PAREMPI results, indicating that reducing precursor gases can effectively lower contribution of traffic to ambient PM levels. Generally, three technologies are identified as having the potential to reduce secondary aerosol formation from transport sectors:

- Advanced exhaust aftertreatment systems (EATS); reducing NMOG, NO_x, NH_3, and particulate matter emissions; however, performance during cold starts needs attention.
- Fuel composition; Limits for fuel monoaromatic and sulfur contents.
- Improved engine and turbine technologies.
- Robust, portable measurement systems to gain reliable data on the SecA emissions and toxicity in real-world conditions. Standardization of testing protocols will be crucial for reliable data.

In road transport, although tightening emission standards have reduced car emissions, there is still room for improvement, particularly during cold-starts, and when PHEVs switch from battery to combustion engine. Among SecA precursors, NO_x emissions are directly regulated, and SO_2 is indirectly controlled through fuel standards. Organic monoaromatic precursor gases could be limited via fuel standards. NH_3 remain unregulated for passenger cars in Europe, highlighting regulatory gap with implications for SecA formation. Further research is essential to fully understand the complex dynamics of SecA formation in real-world driving conditions.

Marine transport: The results emphasize that stringent regulations for sulfur and NO_x emissions in wider regions than current emission control areas would reduce SecA formation in shipping. Additionally, including NH_3 emissions in regulations would mitigate ammonia slip from SCR and ammonia-fueled marine engines. As the sector moves toward cleaner fuels and utilization of EAT systems, addressing all major pollutants would improve air quality and enable meeting international environmental targets.

Aviation transport: These findings highlight the unique challenges faced by the aviation sector in reducing total PM and SecA precursor emissions. While other transport modes benefit from advanced EAT systems, aviation must rely on fuel and engine innovations. In this respect, low-aromatic or sulfur-free SAFs would mitigate environmental impact of aviation.

The work done in PAREMPI clearly indicated that PM_{aged} data from all sectors is not sufficient and more measurements are needed, which target can be alleviated by developed robust measurement systems for both SecA and toxicological analysis. Findings from toxicological tests highlight the need for further systematical in-depth toxicological analysis in real-world conditions and need to address the unregulated pollutants. Also, the emission measurements must be planned to support toxicological studies through appropriate exposure conditions and sample handling and scheduling to enable the representative toxicological results.

Innovations to reduce PM_{aged} emissions may be underestimated, for example, if EAT systems would be invented for aviation sector, or some transport sectors would move to electricity faster than thought. Additionally, SecA prevention could be enhanced by efficient use of tools, such as modelling (including ePMI), PAREMPI databases and robust measurement systems for SecA and toxicity.

PAREMPI Deliverables and Publications

Deliverables:

PAREMPI Deliverable D2.2, Rönkkö et al., Measurement results of emissions, secondary aerosol forming potential and methodology, Horizon Europe project PAREMPI, Available https://parempi.eu/, 2025.

PAREMPI Deliverable D2.3, Aakko-Saksa et al., Evaluation of robust measurement methods for estimating secondary aerosol mass, Horizon Europe project PAREMPI, Available https://parempi.eu/, 2025.

PAREMPI Deliverable D3.1, Roldin et al., Aerosol dynamics during exhaust dilution and cooling, Horizon Europe project PAREMPI, available at https://parempi.eu/, 2024.

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PAREMPI Deliverable D3.3, Roldin et al., European and global scale estimates of the vehicle fleet contribution to organic and inorganic PM2.5 and nano-particle concentrations during all seasons, Horizon Europe project PAREMPI, Available https://parempi.eu/, 2024.

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Conference abstracts

Large number of conference abstracts (>40) has been published based on PAREMPI results. The abstracts are listed in https://parempi.eu/research/publications/ .

Appendices

Appendix 1. Reference value explanations

Appendix 2. List of analysed PAH compounds

Appendix 3. Remarks on robust measurement system

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Appendix 1. Reference values for Section 3.1

Road transport

Reference values for emissions from road transport are based on emission standards. Reference value for the fuel originating SO_2 emission is based on 10 mg/kg fuel sulfur content (approx. SO_2 0.46 mg/MJ). PM_{aged} reference values are from PAREMPI **D5.2,** based on the 1st PAREMPI database version.

Emission standard	**HC**	**NMHC**	**NO_x**	**NH_3**	**PM**	**$PN_{nv}23$**
	mg/km	**mg/km**	**mg/km**	**mg/km**	**mg/km**	**1/km**
Car G-Euro 7	100	68	60	-	4.5	
Car D-Euro 7	-	-	80	-	4.5	
Car - Euro 6						6.00E+11
	mg/kWh	**mg/kWh**	**mg/kWh**	**mg/kWh**	**mg/kWh**	**1/kWh**
HD Euro VII*	-	105	260	85	8	
HD Euro VI						8.00E+11

* RDE, except PM transient **Diesel car, Euro 7: NMHC value is NO_x+HC minus NO_x limit

Marine transport

Reference values for **gaseous emissions** from marine transport are based on Aakko-Saksa et al. (2023), Table S4. EFs for alternative fuels or exhaust aftertreatment are not included as reference values. For ammonia emissions, NH_3 emissions downstream SCR are scarce; one reference reporting as follows (Aakko-Saksa et al. 2023): *"For example, in a study on-board a ship, NH_3 emissions were close to the detection limit downstream of the SCR (NH_3 < 0.026 g/kWh), and below the detection limit after an SCR and scrubber combination. NH_3 is water-soluble and may dissolve in scrubber water."*

Reference values for **$PN_{nv}23$** are from Aakko-Saksa et al. (2023): "*For marine MSD diesel engines, with or without exhaust aftertreatment, using residual (0.7 and 2.2%S) or distillate fuels, PNnv > 23 emissions have been reported from 0.6 × 10^{14} 1/kWh to 3.5 × 10^{14} 1/kWh (Fig. 6d)*."

PM_{aged} reference values are from PAREMPI D5.2, based on the 1st PAREMPI database version.

Aviation

For aviation, reference values for **gaseous emissions** are from M. Winther and K. Rypdal, "Aviation (1.A.3.a, 1.A.5.b)," EMEP/EEA air pollutant emission inventory guidebook 2023," 2023.

PM_{aged} reference values are from PAREMPI D5.2, based on the 1st PAREMPI database version. For aviation, literature referred to secondary organic and/or inorganic aerosols, without black carbon portion of PM.

Based on ICAO data for LEAP engines, the $PN_{nv}10$ variation is large, between 1.11 × 10^{15} #/MJ for lean burn modern engines to 5.33 × 10^{17} #/MJ for old engines still on production. Reference value selected is for lean burn modern engines, since also for the other transport sectors, references are for clean technologies.

Appendix 2. List of analyzed PAH compounds

List of analyzed PAH compounds, including Nitro-PAHs, Oxy-PAHs and DBTs. Marked with * 16 US EPA PAHs.

35 PAHs

Light PAHs

naphthalene*
2-methylnaphthalene
1-methylnaphthalene
biphenyl
2,3-diMeNaphthalene
acenaphthylene*
acenaphthene*
2,3,5-triMeNaphthalene
fluorene*
1-methylfluorene
phenanthrene*
anthracene*
2-methylphenanthrene
3-methylphenanthrene
1-methylphenanthrene
1-methylanthracene
2-phenylnaphthalene

Intermediate PAHs

fluoranthene*
pyrene*
1-methylfluoranthene
retene
1-methylpyrene

Heavy PAHs

benzo(a)anthracene*
chrysene*
2-methylchrysene
5-methylchrysene
benzo(b)fluoranthene*
benzo(k)fluoranthene*
benzo(e)pyrene
benzo(a)pyrene*
perylene
indeno(1,2,3-c,d)pyrene*
dibenzo(a,h)anthracene*
benzo(g,h,i)perylene*
coronene

Nitro-PAHs

1-Nitronaphthalene
2-Nitronapthalene
1,5 dinitronapthalene
5-nitro acenapththene
2-Nitrofluorene
9-Nitroanthracene
9-Nitrophenanthrene
4-Nitropyrene
3-Nitrofluoranthene
1-Nitropyrene
2-Nitropyrene
7-Nitrobenz[a]anthracene
6-Nitrochrysene
3-Nitrobenzanthrone
1,3-Dinitropyrene
1,6-Dinitropyrene
1,8-Dinitropyrene
6-Nitrobenzo[a]pyrene
3-Nitroperylene
7-N-diBz(a,h)anthracene

Oxy-PAHs

Napthalene-1-aldehyde
2-Naphthaldehyde
9-Fluorenone
9,10 Anthraquinone
1,4 Anthraquinone
Phenanthrene-9-aldehyde
Benzo[a]fluorenone
Benzo[b]fluorenone
Benzanthrone
Benz[a]anthracene-7,12-dione

DBTs

dibenzothiophene
2-methyldibenzothiophene
1-methyldibenzothiophene
4-methyldibenzothiophene
2,8-diMe-dibenzothiophene
2,4,7-triMe-dibenzothiophene

Appendix 3. Remarks on the evaluation of the robust measurement methods for SecA.

Table: Remarks on the robust measurement methods for SecA (see section 6.4 and D2.3).

Parameters	Remarks
Sampling of raw exhaust	• Follows general principles in the emission standards. • In-house sampling system used in PAREMPI to allow semi-trailer setup for trucks. • Exhaust flow meter (EFM) and CAN data corresponded well in PAREMPI. • Examples of sampling systems recommended to be collected in the guidelines.
Priority gases	• PEMS-FTIRs suitable for RDE measurements; measure many gases simultaneously including aromatics when these are included in the recipes/methods; Interferences to be considered. • Requirements for the PEMS-FTIRs (and other principles) need to be defined, including recommended detection limits and measurement ranges. • Evaluation of capability of fuel monoaromatic content as proxy for exhaust monoaromatic precursors recommended.
Diluters	• Porous tube (PTD), ejector diluters (ED) and eDiluters used in PAREMPI. EDs and eDiluters are relatively robust. Requirements for diluters recommended to be defined. • Purified dilution air essential before OFR. Cleanliness requirements needed. • Adjustable dilution ratios needed for varying sources.
OFR	• Portable OFR was used alongside PAM in PAREMPI. Portable OFR can offer robust adjustment and control. • Settings (photo-chemical age) recommended to be defined. Reliable measurement of NO_x at ppm level needed.
PM_{aged}	• Many devices, including AMS, BC instruments and particle size-distributed (PSD) number measuring instruments used in PAREMPI. • Recommendation for PN to PM conversion based on PSD for robust system. Many PDS devices are available. In PAREMPI, ELPI+ used provides PN and PM mass in output file. Recommendations for specifications for the PSD devices needed. • Particle density assumed negligible for PM_{aged} conversion for the instrument chosen.
Emission factors (EF)	• Recommended PM_{aged} EF unit is mass per MJ to harmonize EFs over sector and to avoid need for exhaust flow measurement.