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Particle lung deposited surface area ($LDSA^{al}$) size distributions in different urban environments and geographical regions: Towards understanding of the $PM_{2.5}$ dose–response

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ABSTRACT

Recent studies indicate that monitoring only fine particulate matter ($PM_{2.5}$) may not be enough to understand and tackle the health risk caused by particulate pollution. Health effects per unit $PM_{2.5}$ seem to increase in countries with low $PM_{2.5}$, but also near local pollution sources (e.g., traffic) within cities. The aim of this study is to understand the differences in the characteristics of lung-depositing particles in different geographical regions and urban environments. Particle lung deposited surface area ($LDSA^{al}$) concentrations and size distributions, along with $PM_{2.5}$, were compared with ambient measurement data from Finland, Germany, Czechia, Chile, and India, covering traffic sites, residential areas, airports, shipping, and industrial sites. In Finland (low $PM_{2.5}$), $LDSA^{al}$ size distributions depended significantly on the urban environment and were mainly attributable to ultrafine particles (<100 nm). In Central Europe (moderate $PM_{2.5}$), $LDSA^{al}$ was also dependent on the urban environment, but furthermore heavily influenced by the regional aerosol. In Chile and India (high $PM_{2.5}$), $LDSA^{al}$ was mostly contributed by the regional aerosol despite that the measurements were done at busy traffic sites. The results indicate that the characteristics of lung-depositing particles vary significantly both within cities and between geographical regions. In addition, ratio between $LDSA^{al}$ and $PM_{2.5}$ depended notably on the environment and the country, suggesting that $LDSA^{al}$ exposure per unit $PM_{2.5}$ may be multiple times higher in areas having low $PM_{2.5}$ compared to areas with continuously high $PM_{2.5}$. These findings may partly explain why $PM_{2.5}$ seems more toxic near local pollution sources and in areas with low $PM_{2.5}$. Furthermore, performance of a typical sensor based $LDSA^{al}$ measurement is discussed and a new $LDSA^{al}_{2.5}$ notation indicating deposition region and particle size range is introduced. Overall, the study emphasizes the need for country-specific emission mitigation strategies, and the potential of $LDSA^{al}$ concentration as a health-relevant pollution metric.

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1. Introduction

Outdoor air pollution is known to be one of the leading risk factors for premature deaths globally (IHME, 2019; Ritchie and Roser, 2019). Especially the mass concentration of ambient fine particles ($PM_{2.5}$), i.e., particles with a diameter smaller than 2.5 μm, is strongly associated with premature deaths (e.g., Dockery et al., 1993; Burnett et al., 2014; Vodonos et al., 2018). For example, ambient $PM_{2.5}$ has been linked to 3.3–10.2 million premature deaths per year (Lelieveld et al., 2015, Cohen et al., 2017, Vohra et al., 2021). Furthermore, fine particles are connected to cardiopulmonary and neurological diseases, and other severe diseases, e.g., lung cancer (Pope et al., 2002; Raaschou-Nielsen et al., 2013, Power et al., 2016). The World Health Organization's (WHO) global air quality guidelines from 2021 highlight that severe health effects associated with $PM_{2.5}$ are observed at levels that were previously considered low and, consequently, the recommendation for an annual mean $PM_{2.5}$ was lowered from 10 $\mu g/m^3$ to 5 $\mu g/m^3$ (WHO, 2021).

Although it is generally known that the concentration of fine particles is associated with major health problems world-wide, the contributing attributes for these effects are still not fully understood. It is not yet known, how to measure and reduce the health risk of fine particles most efficiently. Usually, the health effects are associated with the mentioned $PM_{2.5,}$ which is also the most measured and reported metric for particles. However, different studies have reported notably varying estimates for premature deaths (e.g., 3.3–10.2 million per year), showing a major uncertainty relating to the health effects of $PM_{2.5}$. Furthermore, the mortality rate from fine particle pollution as a function of annual $PM_{2.5}$ concentration vary significantly depending on the country and the continent (IHME, 2019; Li et al., 2019; Ritchie and Roser, 2019), emphasizing the importance to understand to what extent local characteristics of ambient fine particles explain this variation. Also, it has been suggested that dose–response curve between the health effects and $PM_{2.5}$ and is steeper in areas with low $PM_{2.5}$ and near local pollution sources, e.g., traffic (e.g., Vodonos et al., 2018; Segersson et al., 2021; Strak et al., 2021). All these observations indicate that measurement of $PM_{2.5}$ alone is not adequate to determine the health risk caused by fine particles.

Ambient fine particle concentrations can be measured also with other metrics than $PM_{2.5}$. For example, WHO has recently recommended starting systematic measurements of ultrafine particles (particles <100 nm) and ambient black carbon (BC) in the new air quality guidelines (WHO, 2021). Another metric to measure the potential health impacts of particulate pollution is $LDSA^{al}$, i.e., lung deposited surface area of particles. $LDSA^{al}$ measures the surface area of particles which deposit in the alveoli of the human respiratory tract. The interaction between the pulmonary circulation and the respiration occurs in the alveoli and, therefore, particles entering the alveoli can possibly end up in the blood circulation and be transported into other organs, e.g., the human brain, placenta, and heart (e.g., Heusinkveld et al., 2016; Bové et al., 2019; Calderón-Garcidueñas et al., 2019). The considered metric is the surface area which has been suggested to be more relevant in terms of health effects with nanoparticles than the mass or number (Brown et al., 2001, Oberdorster et al., 2005; Schmid and Stoeger, 2016). Previous studies have indicated $LDSA^{al}$ to have potentially stronger associations with mortality (Hennig et al., 2018, considers also tracheobronchial deposition, i.e., $LDSA^{al,tb}$), reduced lung function (Patel et al., 2018) and subclinical atherosclerosis (Aguilera et al., 2016) than $PM_{2.5}$ or PM_{10}, emphasizing its potential as a relevant metric in health effect studies, although not enough data is available yet for definitive conclusions. Furthermore, $LDSA^{al}$ size distributions provide important information about the possible sources and composition of particles that cause lung-exposure, i.e., having potential effects on health. In general, $LDSA^{al}$ is a relatively easy metric to measure (e.g., Fissan et al., 2006; Fierz et al., 2014), highlighting its potential in air quality monitoring.

To understand the health risk of particulate pollution properly, physical, and chemical characteristics of local particulate matter should be considered. For example, toxicity of inhaled particles depends on their chemical composition and oxidative potential (e.g., Bates et al., 2019), and studies indicate that e.g., BC is more robustly connected with negative health effects than $PM_{2.5}$ in general (Janssen et al., 2011). Furthermore, previous studies have shown that local pollution sources (e.g., traffic, aviation, shipping, and wood combustion) contribute significantly to concentrations of ultrafine particles (e.g., González et al., 2011; Stacey, 2019; Kuittinen et al., 2021, Damayanti et al., 2023, Lepistö et al., 2023), which are also the most likely ones to enter the lung alveoli when inhaled (ICRP, 1994). Hence, ultrafine particles can carry high concentrations of surface-bound components into the human respiratory system (Kwon et al., 2020) and cause high $LDSA^{al}$ exposure (Lepistö et al., 2023). According to a review study by Ohlwein et al. (2019), ultrafine particles are linked to short-term effects on human health such as changes in inflammatory status and cardiovascular conditions. Either ultrafine particles or chemical composition cannot be observed with the common $PM_{2.5}$ measurement. On the other hand, $PM_{2.5}$ emphasizes the role of larger accumulation mode (100 nm–1 μm) particles which are usually more aged and oxidized as well as often detected in regional background aerosol and during long range transported (LRT) pollution episodes (e.g., Leoni et al., 2018, Teinilä et al., 2022, Trechera et al., 2023). Thus, equal $PM_{2.5}$ concentrations in different locations can consist of different combinations of particle sizes, fraction of ultrafine particles, and chemical composition, which should be considered when analysing the relationship between $PM_{2.5}$ and the health effects.

The composition of $PM_{2.5}$ is especially important when considering the recent developments of air quality in Europe. During the last few decades, $PM_{2.5}$ concentrations have decreased notably in Europe (IHME, 2019; Ritchie and Roser, 2019, de Jesus et al., 2020). Especially in the Northern Europe, the concentrations are low and the yearly averaged $PM_{2.5}$ is close to the WHO's recommendation of 5 $\mu g/m^3$. Despite the low concentrations of $PM_{2.5}$, ultrafine particle concentrations can still be high (e.g., de Jesus et al., 2019, Lepistö et al., 2023). Also, the number of the smallest ultrafine particles (<30 nm) originated from traffic has not consistently decreased alike the concentrations of larger particles during recent years (Damayanti et al., 2023). In addition, studies have indicated varying toxicity of combustion emissions depending on the source, emphasizing the toxicity of traffic emissions in comparison e.g., with biomass or coal combustion (Park et al., 2018). Therefore, near-source exposure to local pollution sources and ultrafine particles may have important effects on public health in Europe, which may not be well recognized only with $PM_{2.5}$. Due to the relatively low $PM_{2.5}$ levels in Europe, the relative exposure to local pollutants in comparison with exposure to regional aerosol is likely higher in Europe than in regions with constantly high regional $PM_{2.5}$. This difference with relative exposures could partly explain why equal $PM_{2.5}$ is related to more premature deaths in Europe than e.g., in Asia (IHME, 2019; Ritchie and Roser, 2019; Li et al., 2019;) and why dose–response curves of $PM_{2.5}$ are steeper in areas with low $PM_{2.5}$ (e.g., Vodonos et al., 2018). Thus, when tackling the problem of urban air quality, it is important to understand the effects of local pollution sources and near-source exposure as well as the differences between different urban environments and geographical regions to guide decisions on the priorities of emission source control.

The aim of this study is to compare $LDSA^{al}$ concentrations and size distributions, and their link with $PM_{2.5}$, in various urban environments in different countries and continents. There are only limited number of previous studies reporting $LDSA^{al}$ size distributions in different urban sites and countries, thus the potentially health-relevant differences in the characteristics of lung-depositing particles are not well understood currently. The experimental ambient data was measured in Finland, Germany, Czechia, Chile, and India, covering urban traffic sites, highways, detached-housing residential areas, airports, shipping, and industrial sites. The observed differences between the locations and their significance in terms of health effects are then discussed with an

epidemiological study point-of-view. Finally, a new $LDSA^{al}_{2.5}$ notation that indicates deposition region and particle size range is introduced to enable more reliable comparison of $LDSA^{al}$ in different locations, helping the future analyses of possible health impacts and trends of $LDSA^{al}$ around the world.

2. Materials and methods

2.1. Measurement campaigns

This study includes data of eight ambient measurement campaigns conducted in Finland, Germany, Czechia, Chile, and India. The measurements cover urban traffic sites, highways, residential areas, airports, shipping, and industrial sites. The campaigns with the representative urban sites are collected in Table 1, and the studied cities are shown on map in Fig. 1. The urban traffic sites include traffic environments in cities, e.g., street canyons and avenues with traffic light junctions. Busy roads with at least four lanes in total and higher flow of traffic, including motorways, were counted as highway sites. Studied residential areas were detached-housing areas where wood combustion for heating purposes was common. The airport measurements were done next to busy international airports in Helsinki (HEL) and Düsseldorf (DUS) whereas the shipping sites include Helsinki harbour and riverside of Rhine in Düsseldorf. The industrial sites include measurements next to a steel factory (Raahe) and a coffee roastery (Helsinki). The length of the campaigns varied from 1.5 weeks up to 1 month. Campaigns in Finland, Germany, and Czechia were conducted by utilizing the Aerosol and trace-gas mobile laboratory (ATMo-Lab). The ATMo-Lab is a van where the sample is taken in front of the van above the windshield at the height of 2.2 m and then divided for the instruments installed in the back end of the van. Both stationary and driving measurements with the ATMo-Lab were conducted depending on the measurement site. Any results from the campaigns in Tampere, Düsseldorf and Prague have not been published previously. Data from Helsinki 1, Helsinki 2, Raahe, Santiago and Delhi-NCR (National Capital Region) are from measurements presented in Lepistö et al. (2022), Lepistö et al. (2023), Barreira et al. (2023), Gramsch et al. (2020) and Salo et al. (2021), respectively. $LDSA^{al}$ concentrations and size distributions of campaigns Helsinki 1, Helsinki 2, Raahe, and Delhi-NCR (National Capital Region) have been discussed previously in the corresponding publication.

2.1.1. Measurement cities and countries

The campaigns in Finland were conducted in Helsinki, Tampere, and Raahe in 2019–2021. Helsinki is the capital of Finland, and the Helsinki metropolitan area has a population of approximately 1.2 million. The Tampere sub-region is the second largest metropolitan area in Finland with approximately 400 000 inhabitants. Raahe is a small industrial town in Northern Finland with about 24 000 inhabitants. In general, average air quality in Finland is good and yearly averaged population weighted exposure to $PM_{2.5}$ is estimated to be close to the WHO's recommendation of 5 μg/m^3 (IHME, 2019; Ritchie and Roser, 2019). However, local emission sources (e.g., traffic and residential wood combustion) and LRT-episodes can worsen the air quality occasionally (e.g., Pirjola et al., 2017; Luoma et al., 2021).

Both campaigns in Central Europe (Düsseldorf and Prague) were conducted in the spring of 2022. Düsseldorf is the capital of North Rhine-Westphalia, the most populous state in Germany, and has a population of approximately 600 000. The metropolitan area of Düsseldorf, Rhine-Ruhr, has over 11 million inhabitants and includes major industrial cities of Germany such as Dortmund, Duisburg, Essen, and Bochum. Prague is the capital of Czechia with approximately of 1.3 million inhabitants (2.7 million in the metropolitan area). Average pollution levels in Central Europe are higher than in Finland and, in Germany and Czechia, yearly averaged population weighted level of $PM_{2.5}$ has been estimated to be between 10 and 20 μg/m^3 (IHME, 2019; Ritchie and Roser, 2019).

In Chile, the measurements were carried out in Santiago in winter 2013. Santiago is the capital of Chile, and the population is approximately 6 million (7 million in the metropolitan area). The measurements in Delhi-NCR by Salo et al. (2021) were done in Gwal Pahari. Population of Delhi-NCR is approximately 29 million and it is one of the most populated metropolitan areas in the world. Especially in India, outdoor air pollution is a major problem. For example, average $PM_{2.5}$ in a study conducted in Delhi-NCR during the winter of 2018 was over 150 μg/m^3 (Lalchandani et al., 2021), whereas yearly averaged population weighted $PM_{2.5}$ in India has been estimated to be about 90 μg/m^3 (IHME, 2019; Ritchie and Roser, 2019). In Chile, yearly averaged population weighted $PM_{2.5}$ was estimated to be 21 μg/m^3 in 2017. However, during winter, $PM_{2.5}$ can clearly exceed 100 μg/m^3 (Barrazza et al., 2017; Reyes et al., 2021).

2.1.2. Measurement sites

In-detail information of Helsinki 1, Helsinki 2, Raahe, Santiago and Delhi-NCR campaigns can be found in the corresponding publications (Table 1). In Tampere, measurements were carried out in an urban traffic site and on a highway. The urban traffic site located in the city centre next to one of the main streets (Pirkankatu, 61.4987 N, 23.7356 E). The highway measurements were conducted by driving back-and-forth an arterial road (Paasikiventie) and a motorway (Porintie) which both are one of the busiest roads in-and-out the city in Tampere. A map of the measurement locations and campaign conditions in Tampere are summarized in the Supplementary information (Fig. S1, Table S1).

In Düsseldorf, the ATMo-Lab was utilized in stationary measurements in an urban traffic site, and in driving measurements next to the airport, river Rhine, and on a motorway. The urban traffic site located next to an arterial road (Auf˙m Hennekamp, 51.2055 N, 6.7868 E) in a low emission zone, which allows only the use of vehicles that fulfil EURO 4 (or newer) emission standard. The airport measurements were done in a residential area approximately 1 km away from Düsseldorf (DUS) airport, and the highway measurements were done by driving motorways around the airport. River Rhine is a busy cargo route from inner Germany to North Sea via the Netherlands, and the river traffic measurements were done in a residential area on the riverside. In Prague, two different stationary measurement sites for the ATMo-Lab were utilized: an urban traffic site and a busy highway. The urban traffic site was next to a two-lane street having two tramlines in the middle in Vršovice (50.0664 N, 14.4462 E) and the highway site was next to a busy six-lane arterial road in-and-out the city (50.1175 N, 14.4595 E). Maps

Table 1
Measurement campaigns included in this study.

Campaign	Country	Season	Time	Method	Environments	Campaign description
Helsinki 1	Finland	Summer	August 13th–23rd, 2019	ATMo-Lab	Urban traffic, Highway, Shipping	Lepistö et al. (2022)
Tampere	Finland	Spring	April 29th–May 14th, 2020	ATMo-Lab	Urban traffic, highway	This study
Raahe	Finland	Winter	January 25th–February 4th, 2021	ATMo-Lab	Residential area, Industrial	Barreira et al. (2023)
Helsinki 2	Finland	Winter	March 1st–15th, 2021	ATMo-Lab	Urban traffic, Residential area, Airport, Industrial	Lepistö et al. (2023)
Düsseldorf	Germany	Spring	March 8th–23rd, 2022	ATMo-Lab	Urban traffic, Airport, Shipping, Highway	This study
Prague	Czechia	Spring	March 25th–April 3rd, 2022	ATMo-Lab	Urban traffic, Highway	This study
Santiago	Chile	Winter	July 9th–15th, 2013	Stationary site	Urban traffic	Gramsch et al. (2020)
Delhi-NCR	India	Winter	November 16th–December 14th, 2018	Stationary site	Urban traffic	Salo et al. (2021)

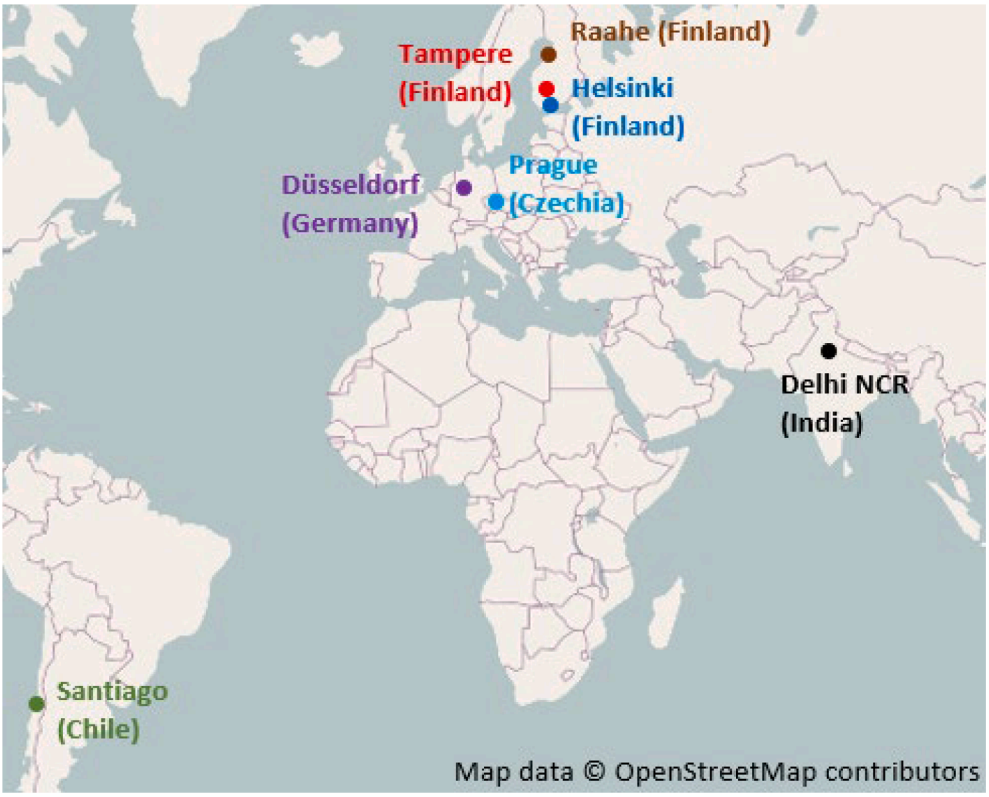

Fig. 1. Cities where the measurements included in this study were conducted.

of the locations and campaign conditions are collected in Fig. S2-3 and Table S2-3.

Due to the relatively short measurement periods, it should be noted that the results represent aerosol only during certain situations and conditions. Therefore, time of the year, role of new technologies, site microenvironments as well as SARS-CoV-2 pandemic etc. have an effect on the measured concentrations. To elaborate the site-dependency, more detailed descriptions of each measurement site are collected in Table S4. The main focus of this study is to compare the effects of varying particle characteristics on $LDSA^{al}$ near different emission sources in the studied locations. Hence, the presented results are selected from periods when the measurement sites were being influenced by the targeted nearby emission sources. For example, only the times when the wind was blowing from the airport were considered in the airport environment data. With campaigns Helsinki 1 (Lepistö et al., 2022), Raahe (Barreira et al., 2023), Helsinki 2 (Lepistö et al., 2023), Chile (Gramsch et al., 2020), and Delhi-NCR (Salo et al., 2021) the utilized data is based either on the criteria in the corresponding publication or data from measurements conducted between 6 am and 6 pm (Table S5). The criteria for chosen periods in Tampere, Düsseldorf, Prague campaigns followed similar principles, as summarized in Table S6.

2.2. $LDSA^{al}$ measurement

Generally, the human respiratory tract is divided into three regions: head airways, tracheobronchial and alveolar. The region where inhaled particles deposit depends especially on the particle size (ICRP, 1994). Deposition efficiencies of spherical particles with the unit density (1 g/cm^3) in the human respiratory tract are presented in Fig. 2. $LDSA^{al}$ refers to particle surface area deposition in the alveolar region which is considered crucial in terms of health effects as interactions between the pulmonary circulation and the respiration occur there. $LDSA^{al}$ as a metric indicates the deposited surface area of particles per cubic centimetre of inhaled air ($\mu m^2/cm^3$). The surface area is considered to be biologically the most effective dose metric for acute nanoparticle toxicity in the human lung (Schmid and Stoeger, 2016). Also, larger surface area allows particles to carry more condensed and toxic material into the lungs. For example, particle coating and aging has been observed to increase soot particle toxicity (Hakkarainen et al., 2022). In addition to the health-relevance, surface area deposition in the alveolar region is also reasonably easy to estimate with sensor devices such as the Nanoparticle surface area monitor NSAM (Fissan et al., 2006), Partector (Fierz et al., 2014), and Pegasor PPS-M (Rostedt et al., 2014), which has increased the popularity of $LDSA^{al}$ as tool for particulate pollution monitoring. However, it should be noted that particle deposition in the other regions likely has effects on human health as well. For example, particles may possibly enter the human brains directly through the olfactory nerve (Maher et al., 2016). The focus in this study is on alveolar deposition ($LDSA^{al}$) as it has become, according to the authors' impression, the most commonly reported and studied method to

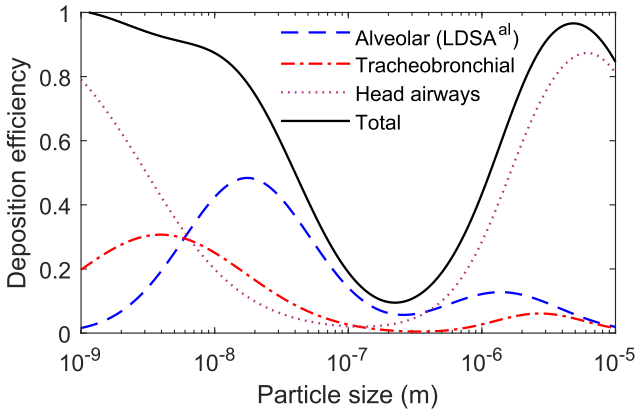

Fig. 2. Particle deposition efficiency in the human respiratory tract as a function of particle size according to ICRP (1994) and Hinds (1999).

measure lung-exposure in ambient measurement studies (see comparisons e.g., by Reche et al., 2015; Kuula et al., 2020).

In addition to particle size, deposition efficiencies depend on other properties of inhaled particles, e.g., density and hygroscopicity. Furthermore, deposition efficiencies are individual and influenced e.g., by physical activity, gender, and age. The efficiencies in Fig. 2 are averaged representations of particle deposition in the human respiratory tract (Hinds, 1999). In general, the effects of particle properties and human anatomy or activity are commonly assumed to be negligible in reported $LDSA^{al}$ concentrations to obtain reasonably accurate estimations of average exposure. Also, with the common sensor-based measurement, these assumptions are needed.

2.2.1. Electrical low pressure impactor

In this study, $LDSA^{al}$ concentrations and size distributions were measured with an electrical low pressure impactor (ELPI+, Dekati Oyj, Keskinen et al., 1992, Järvinen et al., 2014). In the ELPI+, sampled aerosol particles are first charged in a unipolar diffusion charger and then classified in a cascade impactor according to their aerodynamic size. The impactor has 14 stages which are connected to electrometers, enabling the measurement of electrical current caused by the charged particles collected onto the impactor stages. Each impactor stage collects particles from different size range and the electrical current as a function of particle size can be measured. $LDSA^{al}$ concentrations and size distributions can then be determined by utilizing stage-specific conversion factors for each impactor stage (Lepistö et al., 2020). The measurement size range of ELPI+ is from 6 nm to 10 μm and the data is obtained with 1 s resolution. The stage-specific conversion factors enable accurate measurement of $LDSA^{al}$ in the whole measurement size range. In addition, the electrical current data can be converted, e.g., to particle number and mass size distributions and concentrations ($PM_{2.5}$). Same ELPI+ unit was used in all the measurement campaigns expect in Santiago. Measured raw current data with different ELPI+ units is known to be well comparable but conversion to total particle number or mass concentration with different units may cause slight uncertainty (e.g., Salo et al., 2019). With $LDSA^{al}$ conversion, the conversion factors from electric current are significantly smaller than with number or mass and, therefore, the uncertainty is minimal with $LDSA^{al}$ measurement.

The stage-specific $LDSA^{al}$ measurement with ELPI+ differs from the method used by the mentioned $LDSA^{al}$ sensors. In general, $LDSA^{al}$ sensors charge sampled particles and then measure the total current caused by all the sampled particles. This total current is then converted to $LDSA^{al}$ concentration with a more-or-less single conversion factor, usually based on the calibration coefficient at 100 nm. With this method, $LDSA^{al}$ can be measured with reasonable accuracy relatively easy. However, the method is accurate only in the size range of about 20 nm–400 nm (e.g., Todea et al., 2015), depending on the device. In this study, general performance of the $LDSA^{al}$ sensors in the studied environments is demonstrated by utilizing a single-factor calibration with the ELPI+ for $LDSA^{al}$ measurement (Lepistö et al., 2020). The single-factor calibration mimics the method with typical $LDSA^{al}$ sensors by measuring the total current of particles collected onto all the impactor stages and then converting the measured current to $LDSA^{al}$ with a single conversion factor of 0.041 $um^2/(cm^3 fA)$, which corresponds to calibration coefficient at 100 nm.

2.3. Data processing

The common approximations of negligible hygroscopic growth in the human lungs and particle effective density of 1 g/cm^3 were done to enable comparison with different studies and sensor-based measurements. The upper limit of particle size range with all the measurement campaigns was chosen to be 2.5 μm to reduce uncertainties related to inertial particle losses in the sampling systems of different campaigns. The presented particle size distributions are shown as a function of the aerodynamic diameter. All the data from different campaigns has been processed and analysed with same principles, including conversions to PN, $LDSA^{al}$ or mass concentrations and size distributions as well as utilization of arithmetic mean, despite possible varying approaches or methods used in the corresponding publications (Table 1). In Sections 3.3 And 3.5, both $LDSA^{al}$ and PM concentrations are calculated separately for size ranges of 0–100 nm, 100–400 nm, and 400 nm–2.5 μm. These were calculated by integrating the size distributions between the ELPI+ stages 1–4, 5–7, and 8–11. The cut-off sizes for these varied slightly with the two different ELPI+ units, being 6 nm, 93.4–95.7 nm, 379–386 nm, 2.38–2.48 μm, respectively. In section 3.4, the presented PM related death rates as a function of annual population-weighted mean $PM_{2.5}$ concentration are based on data from Global Burden of Disease Study (IHME, 2019; Ritchie and Roser, 2019).

Due to varying definitions, measurement methods, and size ranges for $LDSA^{al}$ in the literature, we introduce a new notation to report $LDSA^{al}$ results to improve the comparison of the results with previous

Table 2
Average $LDSA^{al}_{2.5}$ and $PM_{2.5}$ concentrations in the studied environments during the studied periods (Table S5-6).

Country	City	Environment	Year	$LDSA^{al}_{2.5}$ ($\mu m^2/cm^3$)	$PM_{2.5}$ ($\mu g/m^3$)
Finland	Raahe	Residential area	2021	*22.5*	*11.8*
		Industrial	2021	*18.5*	*6.6*
	Tampere	Urban traffic	2020	*12.2*	3.2
		Highway	2020	*47.9*	10.7
	Helsinki	Urban traffic	2019	*35.4*	*13.4*
		Urban traffic	2021	*13.2*	*3.2*
		Highway	2019	*14.4*	*5.6*
		Shipping	2019	*23.0*	*11.2*
		Airport	2021	*29.2*	*5.0*
		Residential area	2021	*22.6*	*9.3*
		Industrial	2021	*32.1*	*7.3*
Germany	Düsseldorf	Urban traffic	2022	*37.7*	*25.0*
		Highway	2022	*44.7*	*17.8*
		Airport	2022	*37.6*	*18.7*
		Shipping	2022	*39.3*	*28.6*
Czechia	Prague	Urban traffic	2022	*35.4*	*23.9*
		Highway	2022	*74.3*	*26.5*
Chile	Santiago	Urban traffic	2013	*330*	*354*
India	Delhi-NCR	Urban traffic	2018	*329*	*268*

(and future) studies. With the new notation, the considered region of deposition is expressed with a superscript and the measured particle size range is shown in the subscript, similarly as with $PM_{2.5}$. For example, $LDSA^{al}_{2.5}$ indicates surface area deposition of particles smaller than 2.5 μm in the lung alveoli.

3. Results and discussion

3.1. $LDSA^{al}_{2.5}$ and $PM_{2.5}$ concentrations

Average $LDSA^{al}_{2.5}$ and $PM_{2.5}$ concentrations in the studied environments during the studied periods (Table S5–6) are collected in Table 2 (deviations in Table S7). In Finland, the $PM_{2.5}$ concentrations during the measurements were relatively low and the average $PM_{2.5}$ values were below 15 μg/m^3, which is WHO's recommended upper limit for short-term (24-hour) concentration. $PM_{2.5}$ concentrations varied notably between the campaigns and the highest $PM_{2.5}$ values cannot be associated only to certain urban environments. Also, $LDSA^{al}_{2.5}$ concentrations varied between the campaigns and locations. The highest $LDSA^{al}_{2.5}$ concentrations were measured on a highway (Tampere) and near an airport (Helsinki) which can be explained with ultrafine particle emissions from road traffic and aviation (Lepistö et al., 2023). Interestingly, the high $LDSA^{al}_{2.5}$ concentrations in Finland were not always seen as high simultaneous $PM_{2.5}$ (discussed in 3.2–3.3). The varying concentrations within same cities (e.g., Tampere) or within same measurement site (e.g., urban traffic in Helsinki) are related to varying source intensity, conditions, and regional aerosol (Table S4, Section 3.2). In general, the measured $LDSA^{al}_{2.5}$ and $PM_{2.5}$ concentrations are typical for Finnish urban environments (e.g., Kuula et al., 2020; Luoma et al., 2021).

In Central Europe, the average $PM_{2.5}$ concentrations were notably higher than the highest values in Finland. On the other hand, $LDSA^{al}_{2.5}$ concentrations were almost equal to the highest observed concentrations in Finland with exception to the highway site in Prague where $LDSA^{al}_{2.5}$ was significantly higher than in all the other studied locations in Europe. The relative variations between different environments were also lower in Central Europe than in Finland for both $LDSA^{al}_{2.5}$ and $PM_{2.5}$. Similarly as in Finland, high $LDSA^{al}_{2.5}$ concentration, e.g., in Prague highway, was not seen as high simultaneous $PM_{2.5}$, which indicates different impacts of nearby emission sources and regional aerosol with the metrics (discussed in 3.2). In comparison with previous studies, both

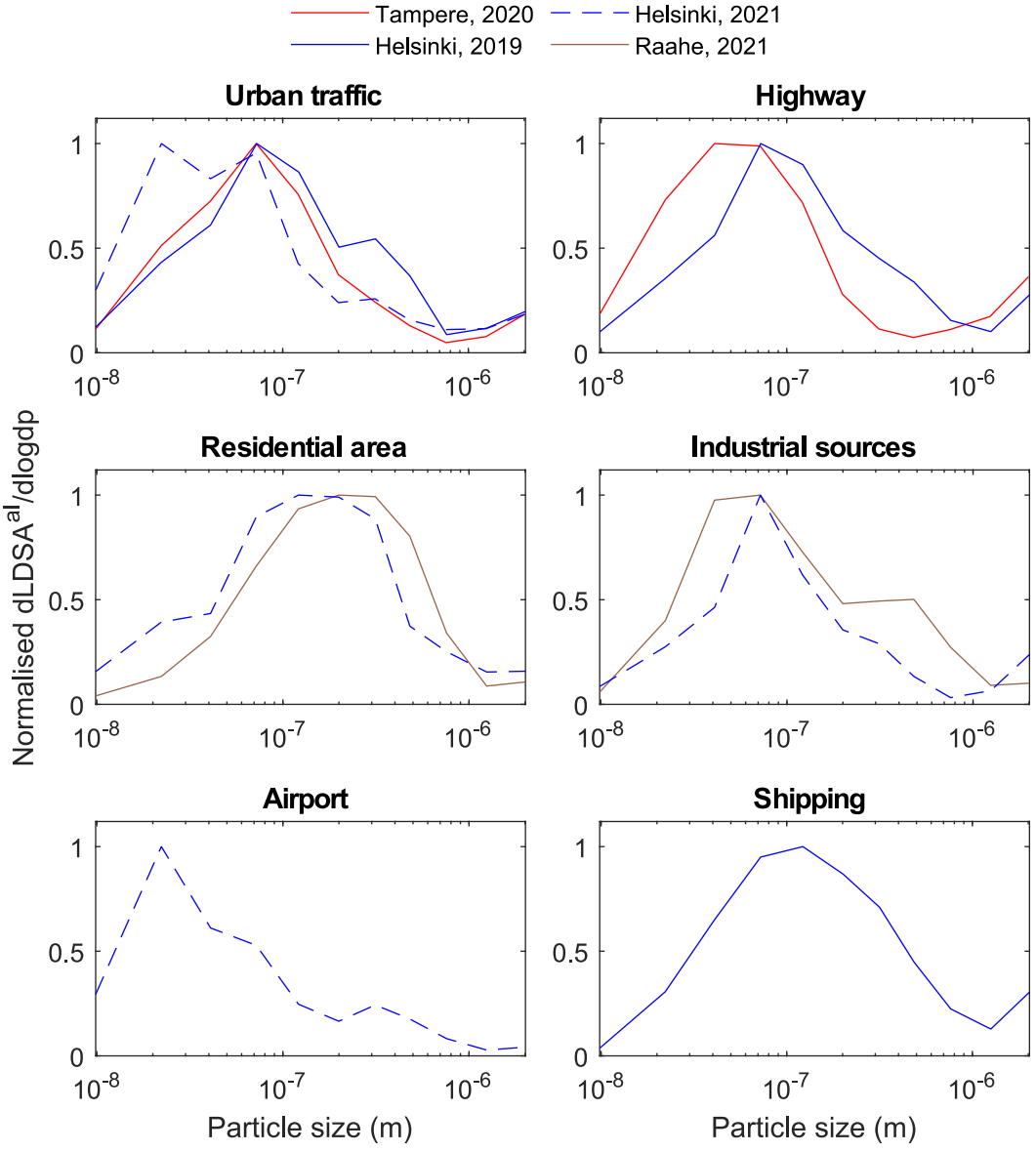

Fig. 3. Normalised average $LDSA^{al}$ size distributions in the studied environments during the studied periods (Table S5-6) in Finland.

$LDSA^{al}$ and $PM_{2.5}$ concentrations were slightly higher than in long-term studies which may relate to seasonal variations. Liu et al. (2023) reported average long-term $LDSA^{al}$ concentrations of 22–32 $\mu m^2/cm^3$ in Germany and Prague in 2017–2019, whereas the long-term averaged $PM_{2.5}$ concentrations have been 10–22 $\mu g/m^3$ in Germany and Czechia (e.g., de Jesus et al., 2020; Liu et al., 2021; Huszar et al., 2021).

In Chile and India, both $LDSA^{al}_{2.5}$ and $PM_{2.5}$ concentrations were significantly higher than in the European cities, emphasizing the difference in exposure to outdoor air pollution in the studied regions. Furthermore, the $LDSA^{al}$ concentrations were by far higher than in previous studies conducted in urban sites (5–164 $\mu m^2/cm^3$) in Europe, USA, and Japan (Cheristanidis et al., 2020). The measured $PM_{2.5}$ concentrations in Chile and India were 9–18 times higher than in the studied sites in Central Europe, whereas $LDSA^{al}_{2.5}$ concentrations were 4–9 times higher, depending on the location, which shows that the relative $LDSA^{al}_{2.5}$ as a function of $PM_{2.5}$ seems to decrease as $PM_{2.5}$ gets higher (discussed in 3.2–3.3). Similar high $PM_{2.5}$ concentrations during wintertime in India have been reported e.g., by Tiwari et al., 2013. In Chile, the measured $PM_{2.5}$ was notably higher than the long-term averages (Barrazza et al., 2017), but similar concentrations of over 300 $\mu g/m^3$ during winter have been reported also in Temuco by Reyes et al. (2021). Hence, measurement campaigns in Chile and India can be considered to represent the most polluted periods of the year.

3.2. $LDSA^{al}$ as a function of particle size

3.2.1. Finland

As seen in Table 2, relationships between $LDSA^{al}_{2.5}$ and $PM_{2.5}$ concentrations varied between the studied environments in Finland which suggests varying particle characteristics in the environments. With $LDSA^{al}$ size distributions, the sizes and characteristics of lung-depositing particles can be analysed. The normalised (by maximum) $LDSA^{al}$ size distributions measured in Finland are collected in Fig. 3. Normalised particle mass size distributions and the absolute size distributions ($LDSA^{al}$ and mass) are provided in Fig. S4-6.

As seen in Fig. 3, each studied environment in Finland had its own typical size range in $LDSA^{al}$ size distribution depending on the nearby emission source. In the airport, $LDSA^{al}$ was clearly dominated by particles smaller than 50 nm whereas, in residential areas with wood combustion, $LDSA^{al}$ was mostly influenced by 100–500 nm particles. With road traffic, $LDSA^{al}$ was mainly contributed by 50–200 nm particles, but the size distributions varied slightly between the campaigns which may indicate differences depending on the season and conditions. For example, weather conditions can affect the dispersion of pollutants and secondary aerosol formation (e.g., Barreira et al., 2021), which change the relative contributions of fresh or aged aerosol. With both shipping and industrial sources, the peak in the distributions is around 100 nm, yet the size distribution is notably wider in the shipping case. As the measurements were done by focusing on the effects of the nearby pollution sources, the observed differences indicate potentially different health effects due to lung exposure of particles from different emission sources. Still, the measured particle mass size distributions did not show notable variation depending on the environment (Fig. S4, discussed also in 3.3). With the exception of residential areas, $LDSA^{al}$ was clearly the most affected by particles smaller than 100 nm, emphasizing the role of ultrafine particle emissions. Also, the peak around 100 nm can often be associated with emissions of BC that contribute strongly to $LDSA^{al}$ (Lepistö et al., 2022).

Variation in the sizes of particles contributing to $LDSA^{al}$ explains the results in Table 2 where the link between $LDSA^{al}_{2.5}$ and $PM_{2.5}$ varied depending on the environment in Finland. The observed major contribution of ultrafine particles in $LDSA^{al}$ in Fig. 3 suggest that the main sources of $LDSA^{al}$ are different than those of $PM_{2.5}$ in Finland. In fact, $PM_{2.5}$ is considerably affected by the regional background concentrations and LRT-pollution in Finland (e.g., Pirjola et al., 2017; Teinilä et al., 2022). In Fig. 4, an example situation of LRT-episode in Helsinki 2 -campaign is shown in comparison with the urban traffic and background site with clean regional background air. The LRT-episode clearly increased $LDSA^{al}$ of 200–800 nm particles, showing a major difference between local and regional aerosol. In Fig. 4, urban traffic, and background site $LDSA^{al}_{2.5}$ concentrations were 13.2 and 4.9 $\mu m^2/cm^3$ on average, respectively, whereas during the LRT-episode average $LDSA^{al}_{2.5}$ was 15.4 $\mu m^2/cm^3$ in the urban background site. On the other hand, $PM_{2.5}$ concentrations were 3.2, 2.4, and 10.1 $\mu g/m^3$, respectively, showing that, during the LRT-episode, $PM_{2.5}$ was over 3 times higher than in the urban traffic site during clean background, whereas the contribution of traffic and LRT-episode on $LDSA^{al}$ was rather similar. In Finland, LRT-episodes are usually observed when arriving air masses have travelled through Central or Eastern Europe (e.g., Pirjola et al., 2017; Salo et al., 2021; Teinilä et al., 2022, Lepistö et al., 2023). The observations emphasize the role of local sources in $LDSA^{al}$, and the role of regional aerosol in $PM_{2.5}$ in Finland, which is discussed in-detail in 3.3.

3.2.2. Central Europe

The normalised $LDSA^{al}$ size distributions in Central Europe are collected in Fig. 5. Normalised particle mass size distributions and the absolute size distributions are provided in Fig. S7-9. As seen in Fig. 5, the

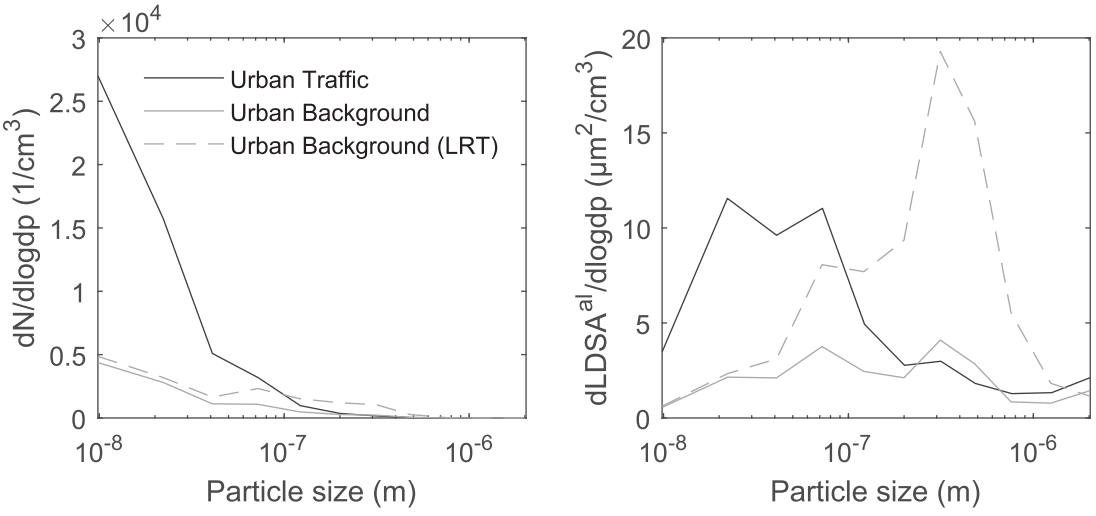

Fig. 4. Effect of long range transported (LRT) pollution event on particle number and $LDSA^{al}$ size distribution in comparison with the urban traffic site in Helsinki 2 campaign during winter 2021.

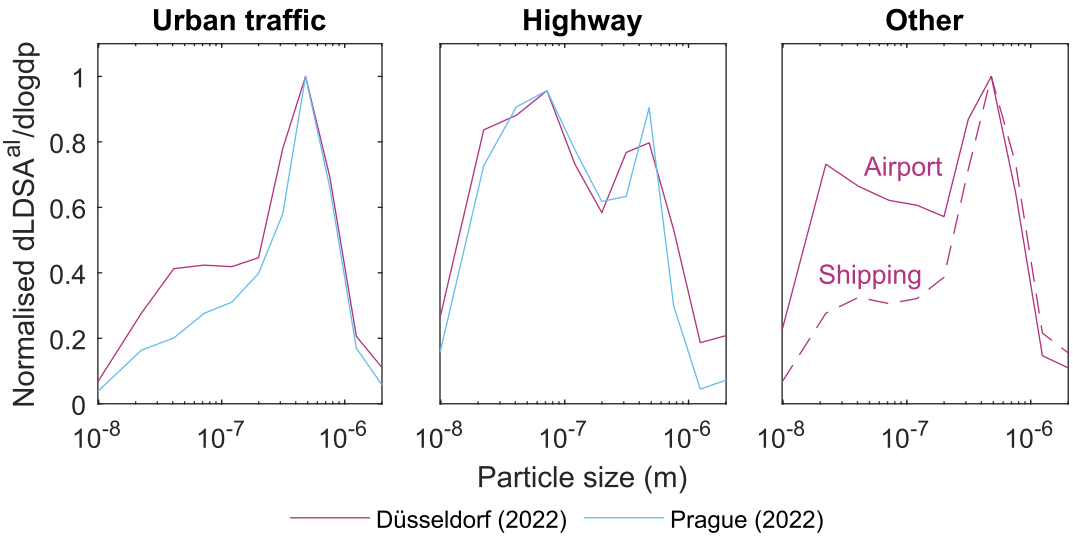

Fig. 5. Normalised LDSA^{al} size distributions in the studied environments during the studied periods (Table S5-6) in Central Europe.

characteristics of LDSA^{al} in Central Europe varied significantly in comparison with Finland. In Central Europe, the LDSA^{al} size distributions had two separate modes, whereas, in Finland, they were mainly one-modal and dominated by particles around 100 nm or smaller. On the other hand, these two separate modes seem to correspond rather well with the peaks caused by the local emission sources and the LRT-episode in Finland (Figs. 3 and 4). In the road traffic sites in Central Europe (Fig. 5), the peak of the smaller particle mode was below 100 nm which agrees well with the results in Finland. Also, in the highways, the contribution of ultrafine particles was higher than in the urban traffic sites which emphasizes the role of traffic in the lung deposition of ultrafine particles as source intensity was notably higher in the highway sites than in the urban traffic sites in Central Europe (Table S4). In the airport, a similar peak below size 50 nm was observed in Düsseldorf as in Finland. However, it should be noted that the airport measurements in Düsseldorf were conducted next to the studied highway (Fig. S2) and, therefore, the highway may have contributed to the result. Still, the peak in the LDSA^{al} size distribution is in smaller particle sizes than with the traffic sites and corresponds well with the results in Finland. Hence, it is likely that the impact of aviation on LDSA^{al} in the studied site is stronger than the effect of the nearby highway. In the shipping site near Rhine, particles originated from the river traffic seemed to be smaller than the ones in the harbour in Finland, which may be related to different fuels, engine types, sizes and regulations with river and marine vessels. In all the studied locations in Central Europe, LDSA^{al} size distributions had a peak with larger particles around 200–800 nm, which agrees well with the mode observed during the LRT-episode in Finland (Fig. 4), indicating that the mode is caused by regional aerosol rather than the nearby local sources. Similarly, as in Finland, the particle mass size distributions did not show notable differences between the studied location in Central Europe (Fig. S7).

The measured size distributions in Central Europe indicate that both local pollution sources and regional aerosol have major contributions on LDSA^{al} exposure. The local pollution sources in Central Europe have similar effects on LDSA^{al} as in Finland but the contribution of regional aerosol is present continuously whereas, in Finland, regional aerosol has considerable effect on LDSA^{al} mainly during LRT-episodes. The consistency in the results of the effects of nearby local sources in Finland and Central Europe highlight potential source-dependency and within city differences in the health effects associated with LDSA^{al} and $PM_{2.5}$. The elevated contribution of regional aerosol in Central Europe may partly be originated e.g., from industrial sources, energy production and residential heating (Pokorná et al., 2018). For example, both Germany and Czechia rely considerably on fossil fuel combustion in energy production

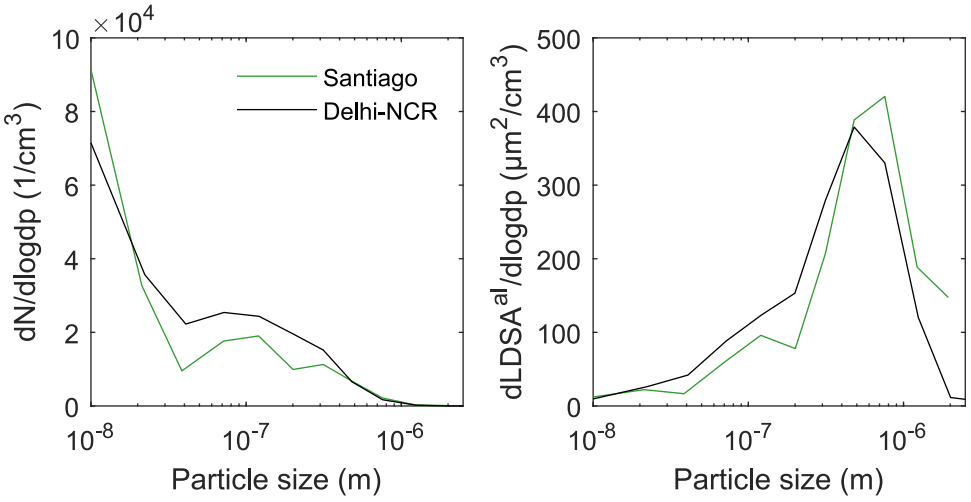

Fig. 6. Particle number and LDSA^{al} size distributions in urban traffic sites in the campaigns conducted in Chile (Gramsch et al., 2020) and India (Salo et al., 2021) during the studied periods (Table S5).

(Eurostat, 2023). Furthermore, coal-based energy-production in Poland has been observed to increase regional background $PM_{2.5}$ (e.g., Rogula-Kozłowska et al., 2014). Also, the dense population in Central Europe is likely part of the explanation.

3.2.3. Chile and India

The particle number and $LDSA^{al}$ size distributions in Chile and India are collected in Fig. 6. Particle mass size distributions and normalised size distributions are provided in Fig. S10-11. As can be seen in Fig. 6, $LDSA^{al}$ size distributions in Chile and India were dominated by 400 nm–1 μm particles, and relative contribution of ultrafine particles was low despite that the measurements were conducted at busy traffic sites. Therefore, in Chile and India, the contribution of regional aerosol on $LDSA^{al}$ clearly exceeded the contribution of local pollution sources, which differs notably from the situation in Finland and Central Europe (Figs. 3 and 5). For example, in India, the high particle concentrations in regional aerosol are affected by the burning of agricultural residue during agricultural seasons (Awasthi et al., 2011). In addition, atmospheric aging of emissions e.g., from traffic and residential sources contribute to increased regional background concentrations. In Chile, the high $PM_{2.5}$ is typically associated with residential biomass combustion and low atmospheric boundary-level height (Reyes et al., 2021). In general, Figs. 3–6 suggest that increased $PM_{2.5}$ indicates increased contribution of regional aerosol on $LDSA^{al}_{2.5}$ and decreased relative contribution of ultrafine particles and local emission sources (discussed in 3.3), which can be seen also with the deviations of the size distributions (Fig. S12). However, it should be noted that particle number size distributions in both Chile and India show high ultrafine particle concentrations, and, therefore, traffic is still a major contributor on $LDSA^{al}$ in both locations despite their relatively low role compared to regional aerosol.

3.2.4. Composition of the lung-depositing particles

The observed varying particle sizes in $LDSA^{al}$ size distributions are important in terms of chemical properties of the lung-depositing particles. In Finland, $LDSA^{al}$ was mainly dominated by local ultrafine particle emissions (Fig. 3). Chemical composition of ultrafine particles is not well understood as it is difficult to detect with mass spectrometer -based measurement. On the other hand, effects of local pollution sources in Finland are usually observed with relatively high BC fraction (16–20 %) of total particulate mass, whereas regional aerosol and LRT-episodes are especially seen with inorganic and organic oxidized aerosol, and fraction of BC is around 10 % or less (Barreira et al., 2021; Teinilä et al., 2022). In Delhi-NCR, previous studies have reported fairly low variability in chemical composition of aerosol measured in different sites (Lalchandani et al., 2021; Shukla et al., 2021), which supports the idea that the effect of nearby local sources is low in comparison with regional aerosol. However, the chemical composition of regional aerosol seems to be different in Delhi-NCR than in Finland as organics cover 28–50 % of total particulate mass (Lalchandani et al., 2021; Shukla et al., 2021), whereas, in Finland, contribution of organics can be 60–75 % in regional aerosol (Teinilä et al., 2022). Oxidized species dominate the organic aerosol composition in regional aerosol in both Finland and India but the increased contribution of local sources in Finland can be observed with decreased fraction of oxidized organic aerosol (Teinilä et al., 2022; Barreira et al., 2023). Therefore, it can be considered that the local sources typically contribute to lung deposition of soot and less-oxidized aerosol, whereas the regional aerosol contributes to lung deposition of oxidized species even though the exact chemical composition cannot be generalized because of different pollution sources, urban environments, and geographical regions.

The varying composition of locally emitted and regional aerosol is important in terms of the health effects. For example, in a study by Jalava et al. (2006) ambient fine particles had lower activity in cytokine production during an LRT-episode than on average in Finland, which was suggested to be due to chemical transformation of the organic fraction during aging processes. Also, in toxicological analysis, decreasing particle size of combustion emissions has been associated with increased toxicity (Hakkarainen et al., 2022). Furthermore, in Delhi (Puthussery et al., 2020), where oxidative potential (OP) of aerosol is mainly caused by aged secondary organic aerosol, the OP per particle mass is lower than e.g., in USA (Puthussery et al., 2018) where OP is also affected by local traffic in addition to secondary organic aerosol. Thus, it is likely that the two different modes observed in the $LDSA^{al}$ size distributions, i.e., local $LDSA^{al}$ and regional $LDSA^{al}$, have varying toxicity, highlighting the role of near-source exposure from local sources in terms of health effects. Moreover, the source-specific results in Figs. 3–6 emphasize the importance of in-detail characterization of emissions from different sources to better understand the potential health effects of particle lung deposition in different urban environments.

3.3. Local and regional $LDSA^{al}$

As observed in section 3.2, $LDSA^{al}$ size distributions consisted of two different modes, which were affected either by local or regional pollution. The effects of nearby local emission sources were seen mainly with ultrafine particles whereas regional aerosol affected $LDSA^{al}$ of particles larger than 400 nm (Figs. 3–6). To analyse the different contributions of local and regional $LDSA^{al}$ in-detail, the measured $LDSA^{al}$ was divided into three different size bins: 0–100 nm, 100–400 nm, and 400 nm–2.5 μm. The 0–100 nm bin covers ultrafine particles and, therefore, can be associated with local emission sources, whereas the 400 nm–2.5 μm bin can be considered to represent the regional aerosol. However, it should be noted that in all cases it is not straightforward to categorize certain pollution source only to a local or regional one based on the particle size, hence the categorization to local and regional aerosol based on the particle size is only an indicative approximation. Especially, the 100–400 nm bin cannot be associated directly to local nor regional sources as it is likely affected by both and, therefore, the results in this section focus on the $LDSA^{al}$ contributed by particles smaller than 100 nm ($LDSA^{al}_{0.1}$) and 2.5 μm ($LDSA^{al}_{2.5}$). Results with the different size bins are shown in Fig. S13 and Table S8.

In Fig. 7, $LDSA^{al}_{2.5}$ and $LDSA^{al}_{0.1}$ (particles smaller than 100 nm) in the studied environments are shown. Despite that $LDSA^{al}_{2.5}$ concentrations in Chile and India were significantly higher than in Finland or Central Europe, the highest $LDSA^{al}_{0.1}$ concentrations in each of the studied regions were rather close to each other. The highest contribution of ultrafine particles on $LDSA^{al}$ was generally associated with either road traffic or aviation.

In Fig. 8, the fraction of $LDSA^{al}_{0.1}$ in total $LDSA^{al}_{2.5}$ is shown and compared with the particle mass concentration. In Finland, 25–73 % of $LDSA^{al}_{2.5}$ was contributed by ultrafine particles whereas only 2–9 % of $PM_{2.5}$ was caused by ultrafine particles, depending on the urban environment. Also, in Central Europe, 22–52 % of $LDSA^{al}_{2.5}$ was caused by ultrafine particles, whereas the corresponding fraction in $PM_{2.5}$ was 1–4 %. Also, as suggested in section 3.2, the fraction of $LDSA^{al}_{2.5}$ contributed by ultrafine particles in Chile and India was low (9–13 %).

The observed differences in Figs. 7 and 8 can also be seen in the association between $LDSA^{al}_{2.5}$ and $PM_{2.5}$. In Fig. 9, the comparison of ratios between $LDSA^{al}_{2.5}$ and $PM_{2.5}$ in all the studied environments is shown. In Finland, the ratio varied from 2.0 to 5.8, showing great dependence on the urban environment and conditions. Especially the role of conditions should be noted as the ratio depends on the $PM_{2.5}$ concentration and, therefore, on the regional aerosol. As the contribution of regional aerosol increased, also the ratio between $LDSA^{al}_{2.5}$ and $PM_{2.5}$ dropped. In Central Europe, the ratio varied from 1.4 to 2.8, and, in Chile and India, from 0.9 to 1.2. Thus, according to the results, $LDSA^{al}_{2.5}$ per unit $PM_{2.5}$ -ratio seem to increase as a function of decreasing $PM_{2.5}$ and the increase is especially steep with low $PM_{2.5}$ -levels (Fig. S14). Also, the $LDSA^{al}_{2.5}$ per unit $PM_{2.5}$ -ratio increases near local ultrafine particle sources, which can be observed also with higher

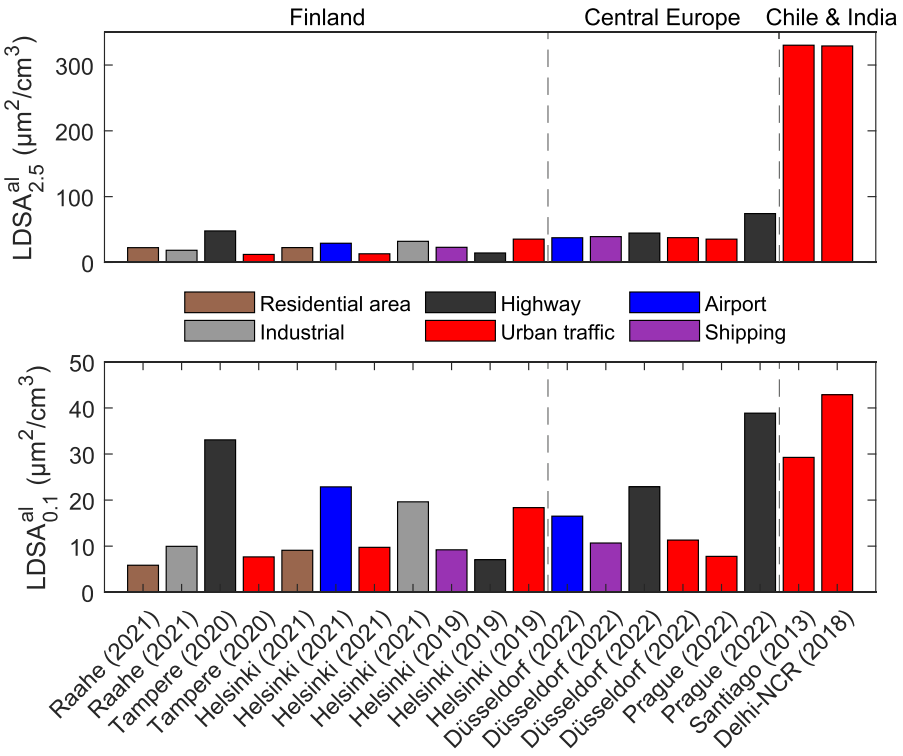

Fig. 7. LDSA^{al} concentrations of particles smaller than 2.5 µm (upper) and 100 nm (below) in the studied environments during the studied periods (Table S5-6). Note varying y-axes.

deviation e.g., due to temporal differences in source-intensity (Table S7). Similarly, the correlation between the metrics reduces near local ultrafine particle sources and in areas with high LDSA$^{al}_{2.5}$/PM$_{2.5}$ -ratio, whereas the correlation gets stronger as the contribution of regional aerosol increases and also near sources that emit larger particles e.g., wood combustion (Table S7, Fig. S15).

To summarize the results in Figs. 7, 8 and 9, it can be considered that in low polluted regions (e.g., Finland) local pollution sources and ultrafine particles may cause relatively high LDSA^{al} exposure, which is difficult to observe by monitoring only PM$_{2.5}$. In high polluted regions (e.g., Chile and India) LDSA^{al} exposure is mainly driven by regional aerosol, which is well observed with PM$_{2.5}$. Thus, relative particle lung deposition per unit PM$_{2.5}$ depends notably on regional aerosol and PM$_{2.5}$ levels, which likely affect the health effects related to PM$_{2.5}$ (discussed in 3.4). The results highlight the importance of monitoring ultrafine particle concentrations as they can cause high LDSA^{al} exposure with only a minimal effect on PM$_{2.5}$ concentration especially in cities.

3.4. *LDSA^{al} and health effects of particles*

The health effects of particulate pollution per unit PM$_{2.5}$ are generally estimated to be similar around the world and the same dose–response curve is used independently regardless of the geographical area. However, the association with premature deaths and PM$_{2.5}$ may significantly depend on the country and geographical region (Li et al., 2019) and, therefore, it may not be reasonable to use the same

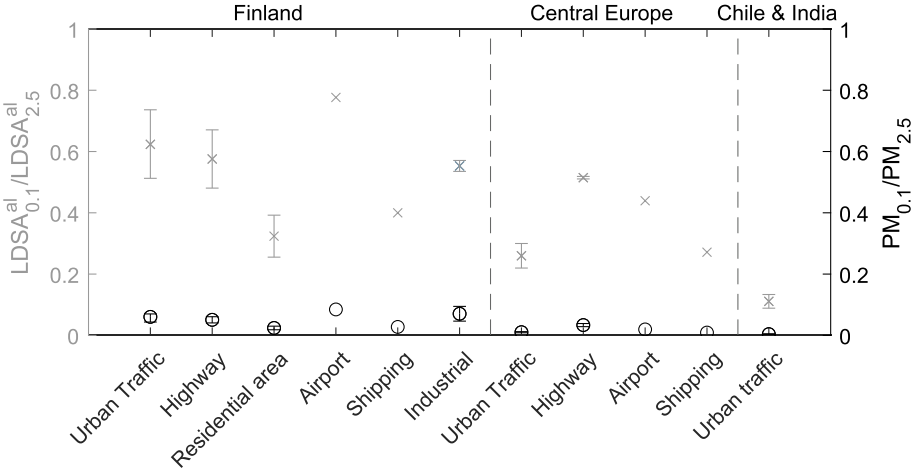

Fig. 8. Fraction of LDSA$^{al}_{2.5}$ and PM$_{2.5}$ contributed by ultrafine particles (<100 nm) in different urban environments in Finland, Central Europe and Chile and India during the studied periods (Table S5-6). Cross and circle signs indicate the average result of all the campaigns conducted in the environment in the certain region and error bars indicate variation between the campaigns. Only a cross/circle sign is shown for results based on single campaigns.

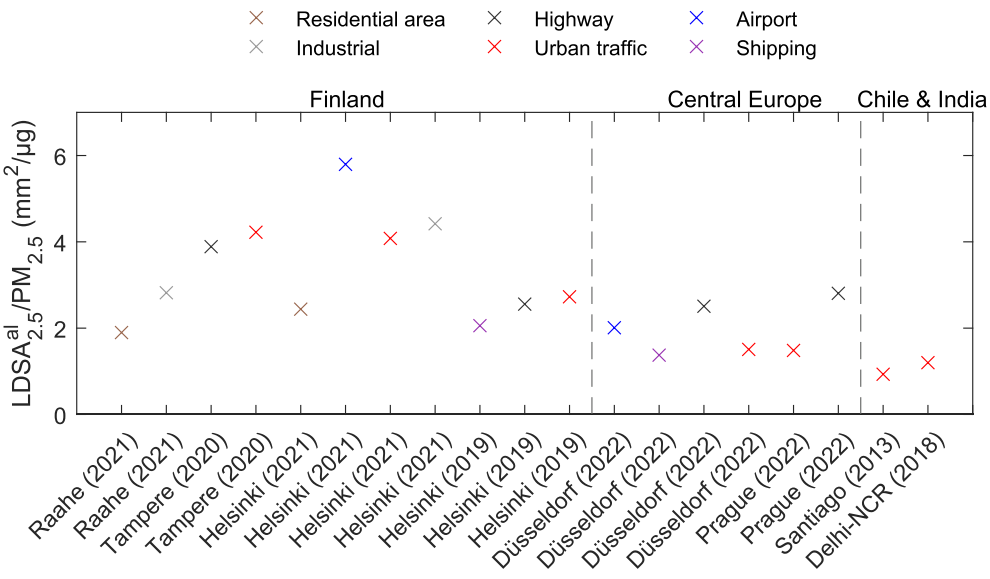

Fig. 9. Ratio between measured $LDSA^{al}_{2.5}$ (µm^2/cm^3) and $PM_{2.5}$ (µg/m^3) in the studied urban environments when the sites were being influenced by the targeted emission source (Table S5-6).

dose–response curve for different regions. To demonstrate this, we used the data of premature deaths linked to PM and an annual population weighted mean $PM_{2.5}$ of different countries (IHME, 2019; Ritchie and Roser, 2019) to compare health effects of $PM_{2.5}$ in different geographical areas. Scatter plots of premature deaths from PM as a function of population weighted $PM_{2.5}$ separately for Europe, Asia, and South America, including calculated linear fits based on the data, are shown in Fig. 10. Furthermore, separate datapoints for Finland, Germany, Czechia, Chile, and India, along with the linear fits for Europe, Asia, and South America, are shown in Fig. S16. It should be noted that as measurement protocols, standards, and locations of $PM_{2.5}$ measurement vary depending on the country, the data should only be considered as indicative. According to our calculations, slope between PM related mortality rate and $PM_{2.5}$ is for example over 4 times higher in Europe than in Asia (Fig. 10). Also, correlation between $PM_{2.5}$ and premature deaths is stronger in Europe (R2 = 0.85) than in Asia (R2 = 0.28) and South America (R2 = 0.43). The stronger correlation may be explained with homogeneity of European countries in terms of air pollution as the annual population weighted mean $PM_{2.5}$ is below 20 µg/m^3 almost in all the countries. The high variation in the results, especially in Asia, emphasize the need to understand the local reasons for the mortality rates caused by PM. According to the data, using the same dose–response curves when estimating health effects of $PM_{2.5}$ in different geographical areas and countries may not be reasonable. However, it should be noted that the mentioned data limitations likely affect the observed variations (e.g., the constant terms of the fits are not zero), especially in Asia and South America.

In Sections 3.2 And 3.3, it was observed that, in Europe, $LDSA^{al}$ was notably more depended on local emission sources and ultrafine particles than by regional aerosol, which dominated $LDSA^{al}$ in Chile and India. Furthermore, the results suggested that the high relative contribution of local pollution sources on lung deposition cannot be well observed by monitoring only $PM_{2.5}$. Now, according to Fig. 10, $PM_{2.5}$ is related to higher mortality rates in Europe than in Asia and South America. Hence, it is plausible that the contribution of local emission sources and ultrafine particles in particle lung deposition, in addition to the discussed potentially increased toxicity of local particle emissions, could partly explain this result. This idea is also supported by previous studies, where health risk per $PM_{2.5}$ increased near local pollution sources and in areas with low $PM_{2.5}$ (e.g., Vodonos et al., 2018; Segersson et al., 2021). In

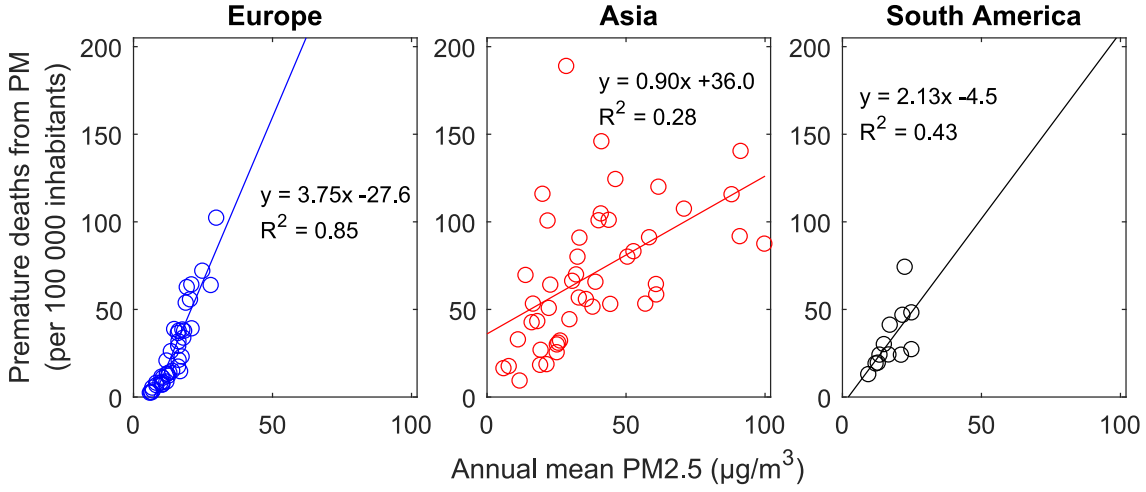

Fig. 10. Association between $PM_{2.5}$ and premature deaths from particulate pollution in Europe, Asia, and South America based on data from Global Burden of Disease Study (IHME, 2019). Each dot represents a separate country. Also, linear fits are applied to the data.

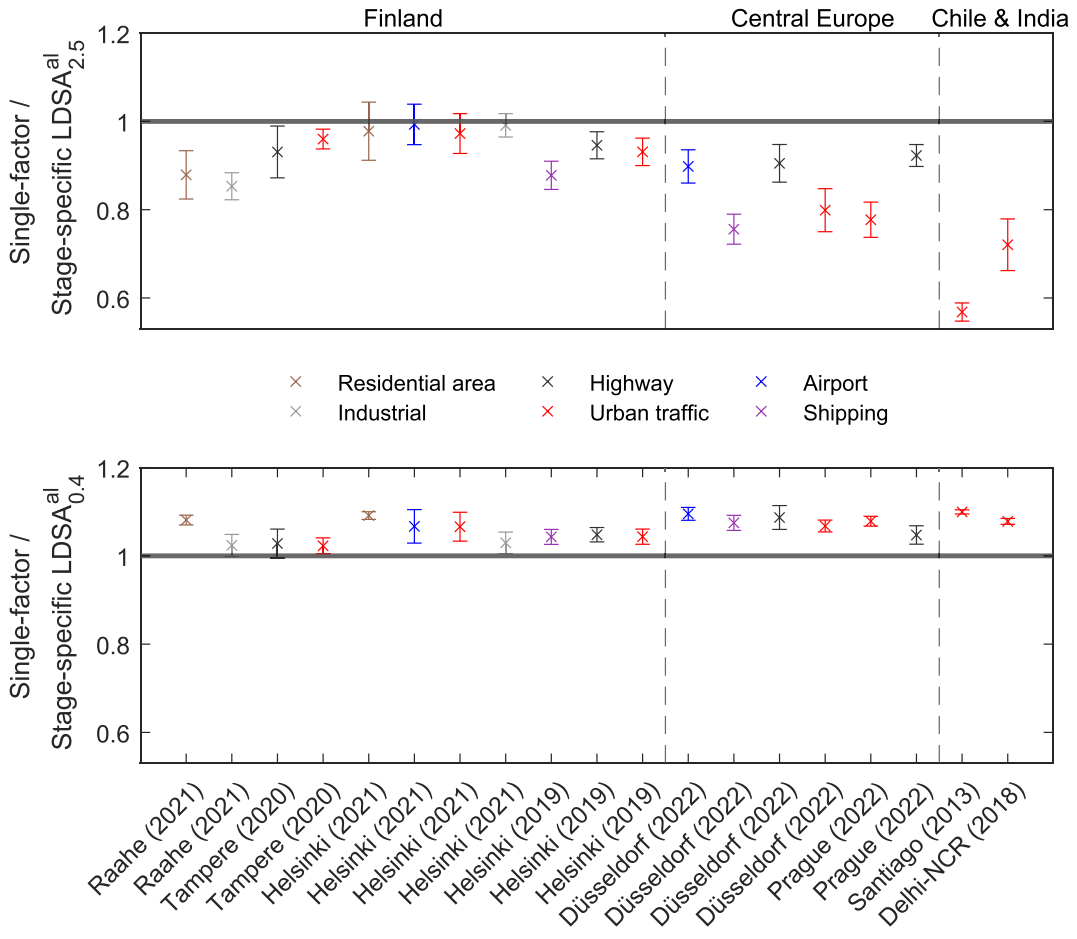

Fig. 11. Performance of the single-factor ELPI+ LDSA^{al} calibration, which mimics the operation of typical LDSA^{al} sensors, against the stage-specific LDSA^{al} calibration utilized in this study with particles smaller than 2.5 μm and 400 nm. The error bars are based on standard deviation.

Chile and India, the contribution of regional aerosol on LDSA^{al} was significantly higher than the contribution of local sources, which indicates that the relative role of ultrafine particles and local sources in $PM_{2.5}$ is also notably lower, which could explain the lower mortality rates as a function of $PM_{2.5}$. Thus, the observed varying $LDSA^{al}_{2.5}$ characteristics between high and low polluted regions may be an important explanation for the toxicity and health effects of $PM_{2.5}$.

The role of local pollution and ultrafine particles in the health effects of $PM_{2.5}$ could also be discussed in terms of the ELAPSE-project, a European project aiming at studying health effects of low levels of air pollution which has been modelled with 100 m × 100 m spatial resolution across Europe. For example, clear associations between not only $PM_{2.5}$, but also with NOx and BC and all-natural causes of death, have been observed in the ELAPSE-studies (Brunekreef et al., 2021; Stafoggia et al., 2022). The associations with NOx and BC likely reflect the effects of ultrafine particles better than $PM_{2.5}$ as both are mainly originated from local pollution, e.g., traffic. The ELAPSE studies could also be mentioned when considering the differences between the countries, as the ELAPSE studies among others suggest that the dose–response curve of $PM_{2.5}$ may not be linear, and that the curve seems to be steeper at the lowest concentrations, even within a low-level area (Vodonos et al., 2018). A similar steep increase of $LDSA^{al}_{2.5}$ per $PM_{2.5}$ -ratios as a function of decreasing $PM_{2.5}$ can be observed in the results of this study (see also Fig. S14), which support the idea that LDSA^{al} and ultrafine particles may have an important role in the increased health effects per unit of $PM_{2.5}$ in areas with low $PM_{2.5}$. However, it should be noted that in the ELAPSE studies, the cohorts with the lowest concentrations came from Sweden and Norway and might not be representative for other populations.

Even though the results with $LDSA^{al}_{2.5}$ per $PM_{2.5}$ -ratios seem to have similar behaviour as the dose–response curve of $PM_{2.5}$ it should be noted that LDSA^{al} does not cover and explain all the possible health impacts of particles. In general, when discussing the increased contribution of ultrafine particles on LDSA^{al}, it should be noted that there is still limited understanding of the connection between ultrafine particles and e.g., mortality (Vallabani et al., 2023). Furthermore, epidemiological studies have suggested that quality of exposure assessment, e.g., due to spatial resolution, is likely explaining some of the observed differences between the countries and associations for near-source and regional air pollution. In ELAPSE, the associations were generally stronger in the cohorts where it was possible to adjust for lifestyle factors than in the administrative cohorts and also, according to Vodonos et al. (2018), studies which controlled for socioeconomic status resulted in higher mortality effect estimates. In addition, Vodonos et al. (2018) suggested that in countries or regions with higher quality of exposure estimates, the observed associations will be stronger. Also, indoor air quality and how people spend time indoor compared to outdoor is likely affecting the relationship between outdoor air pollution and health effects in different geographical regions (e.g., Morawska et al., 2013). All these mentioned factors could be partial explanations for the differences in $PM_{2.5}$ health effects between regions and countries. In addition, the properties of the inhaled aerosol, including chemical and physical characteristics, are known to affect the toxicity of particulate pollution. For example, Park et al. (2018) ranked different emission sources based on the toxicity of $PM_{2.5}$, suggesting that diesel or gasoline exhaust particles are more toxic than e.g., biomass or coal combustion originated particles. In a review study by Nel et al. (2006), it is stated that the ability of generating reactive oxygen species (ROS) and oxidative stress are important factors when considering the toxicity of inhaled nanoparticles. Also, according

to a review by Bates et al. (2019), oxidative potential of aerosol is more strongly associated with acute cardiac and respiratory health end points than PM mass. Therefore, it is well understood that the aerosol properties and composition, and thus the health effects, are depended on the region and country, as discussed also in 3.2.4. However, based on the results, $LDSA^{al}$ should be considered as an additional and potential explaining factor in the health effects of particulate pollution as the varying aerosol characteristics do not only affect the toxicity but also the relative dose of particles in human lungs as a function of $PM_{2.5}$. The observed similarities with the $PM_{2.5}$ dose–response and $LDSA^{al}_{2.5}$ per $PM_{2.5}$ -ratios emphasize the potential of $LDSA^{al}$ in terms of the health effects, being possibly even more relevant than $PM_{2.5}$.

3.5. $LDSA^{al}$ measurement techniques and notation

$LDSA^{al}$ measurement has become more common during the recent years and the results of this study among others have highlighted its potential in terms of particle health effects. However, varying measurement techniques and definitions for $LDSA^{al}$ complicate the comparison of different studies and health effect estimations. To understand the health effects of $LDSA^{al}$ better, it would be important to have better comparability between different studies reporting $LDSA^{al}$. In this study we introduced a new notation for $LDSA^{al}$ measurement, indicating the deposition region and particle size range, to improve the comparison of the reported results with previous and upcoming studies.

As mentioned, typical $LDSA^{al}$ sensors use a method which is reasonably accurate with particles smaller than approximately 400 nm. However, as seen in Sections 3.2 And 3.3, significant fraction of $LDSA^{al}$ can be caused by larger particles, especially in the most polluted regions, indicating that $LDSA^{al}$ results with sensors may not be comparable with measurements that cover a larger particle size range and size distributions. In Fig. 11, the performance of the typical $LDSA^{al}$ sensor method was mimicked with the single-factor ELPI+ $LDSA^{al}$ calibration (Lepistö et al., 2020) and then compared with the stage-specific $LDSA^{al}$ method utilized in this study. The comparison was done separately with particles smaller than 2.5 μm and 0.4 μm. As can be seen, the ratio between the single-factor and the stage-specific calibration varied from 57 % to 99 % when considering particles smaller than 2.5 μm, showing a major difference between the methods depending on the environment, and indicating major uncertainties related to $LDSA^{al}$ sensor measurements in different locations. On the other hand, if only particles smaller than 0.4 μm are considered, the ratio between single-factor and stage-specific methods is 102–110 %. Therefore, when reporting $LDSA^{al}$ results, it would be beneficial to report the size range of $LDSA^{al}$ measurement, similarly as with $PM_{2.5}$, to improve the comparison of different studies conducted in different environments even though device-specific operation principles also affect the comparability of the reported results. The good comparability with particles smaller than 0.4 μm also suggest that $LDSA^{al}$ sensors may be an effective and relatively easy method to monitor the concentrations and potential health effects of particles emitted from local pollution sources as long as the effect of larger particles is taken into account. In addition to particle size, it is important to state the considered respiratory tract in the metric, as there is no strict definition for LDSA. In addition to $LDSA^{al}$ sensors, it should be noted that the principles with size distribution measurements vary depending on the device as well. In general, these differences between different $LDSA^{al}$ sensors and different size distribution measurement methods in terms of $LDSA^{al}$ are not well understood, highlighting the need for future studies. Overall, the results show that particles larger than 400 nm (and even larger than 1 μm) can be major contributors on total $LDSA^{al}$ concentration which should be taken into account in future studies focusing on $LDSA^{al}$.

4. Strengths and limitations

According to the results of this study, $LDSA^{al}$ size distributions and exposure significantly depend on the urban environment and geographical region. Also, the results show that $LDSA^{al}$ concentration per unit $PM_{2.5}$ can be multiple times higher in areas having low $PM_{2.5}$ (e.g., Finland) compared to areas with continuously high $PM_{2.5}$ (e.g., India). In addition, this study shows that local pollution sources and ultrafine particles are important contributors on $LDSA^{al}$, especially in regions with low $PM_{2.5}$. These results are important when considering the health effects associated with $PM_{2.5}$ as they indicate that health effects due to particle lung deposition likely vary in different geographical regions but also within cities due to varying characteristics of lung-depositing particles. These differences are not easily observed with $PM_{2.5}$. Despite that it is generally understood that varying physical and chemical characteristics of the aerosol affect the toxicity and health effects of $PM_{2.5}$, this study shows that, in addition to the toxicological properties, the varying aerosol characteristics also significantly affect the relative lung exposure per unit $PM_{2.5}$ in different regions. The observed similarities with the measured $LDSA^{al}_{2.5}$ per $PM_{2.5}$ -ratios and $PM_{2.5}$ dose–response highlight the potential of $LDSA^{al}$ as a health relevant pollution metric, which could help to estimate the disease burden due to air pollution more accurately. However, it should be noted that definitive conclusions about the health impacts of $LDSA^{al}$ are difficult to draw based on the existing literature. Also, the understanding of the health effects of ultrafine particles is still limited (Vallabani et al., 2023). Thus, $LDSA^{al}$ provides another potential explanation for the varying health effects of particulate pollution, but other explanations such as aerosol chemical composition, oxidative potential as well as differences with populations should not be forgotten.

It is important to note that the study lacks long-term measurement data, which should be considered when interpreting the results. The reported results represent the aerosol only during certain situations and conditions. Also, site microenvironments, varying source-intensities as well as different seasons, SARS-CoV-2 pandemic (Table S4, Section 2.1.2) affect the reported values and complicates the comparison between the sites. Therefore, the reported absolute concentrations (Table 2) should not be generalized to represent long-term averaged pollution levels in the studied sites. The focus of this study is on the effects of different emissions sources and regional aerosol on $LDSA^{al}$, and therefore, the considered data was chosen from periods when the measurement sites were being influenced by the targeted nearby emission sources (Table S5-6). With this approach, the discussed different effects of nearby local sources and regional aerosol as well as the differences between the regions are apparent, especially with the $LDSA^{al}$ size distributions. For example, the results from different urban environments are consistent in different cities and regions. Also, the contribution of regional aerosol is coherent in all the studied regions. Therefore, the typical effects (e.g., contributing particle size) of nearby emission sources on $LDSA^{al}$ as well as the fundamental behaviour of increasing $LDSA^{al}_{2.5}$ per unit $PM_{2.5}$ -ratio as a function of decreasing $PM_{2.5}$ can be derived from the results of this study. Still, it should be noted that the relative contribution of local and regional pollution depends on time and conditions, highlighting the need for future long-term studies, with a large measurement size range of particles, to better understand all the factors contributing to $LDSA^{al}$ in different environments and regions.

The study focuses on the potential health effects relating to the alveolar deposition ($LDSA^{al}$) of particles. It is important to note that particle deposition also in the other respiratory tract regions has potential health impacts. In general, $LDSA^{al}$ as a metric lacks strict definitions and the differences between different measurement methods are not well understood. For example, $LDSA^{al}_{2.5}$ calibration of ELPI+ is based on deposition model by ICRP (1994), whereas some studies have utilized the multiple path particle dosimetry (MPPD) model (e.g., Liu et al., 2023). Also, it is evident that results with common $LDSA^{al}$ sensors can vary significantly in comparison with size distribution measurements (Fig. 11). Therefore, $LDSA^{al}$ -studies include uncertainty with the reported values due to varying methods and initial approximations related

e.g., to particle density and hygroscopicity (discussed in 2.2). On the other hand, one of the main strengths of this study is the consistency of the methods, including the same measurement device and data processing approaches, and, therefore, the comparison between different sites is consistent and reliable. Also, the results highlight the necessity of wide enough measurement particle size range (up to 2.5 μm or even larger) of $LDSA^{al}$ in future studies. Thus, further discussions of $LDSA^{al}$ definitions and measurement principles are needed in the future as $LDSA^{al}$ is a potential metric in terms of health effects and also relatively easy to measure. In this study, the new $LDSA^{al}_{2.5}$ notation was introduced to encourage this discussion and also to improve the comparison of the results of this work with previous and upcoming studies.

5. Conclusions

The results of this study suggest that $LDSA^{al}$ concentrations and size distributions are potential additional explaining factors for the varying health effects of particulate pollution in different urban environments, different geographical regions, and as a function of $PM_{2.5}$. Despite the potential of $LDSA^{al}$ as a health relevant pollution metric, there are varying definitions and measurement principles for $LDSA^{al}$ in literature, which complicates the health effect estimations relating to the metric. For example, as shown by the location-dependent size distributions, the sensor based $LDSA^{al}$ measurement methods may possess considerable uncertainties in heavily polluted environments. Further discussions of $LDSA^{al}$ definitions and measurement methods as well as further investigations of long-term $LDSA^{al}$ characteristics could help to understand to what extent the utilization of $LDSA^{al}$ as a monitored metric could contribute to the understanding the health effects of particulate pollution and the steepness of the $PM_{2.5}$ dose–response function. In all, the results of this study emphasize the importance of location and country specific emission mitigation strategies and, for example, in Europe, focus on local ultrafine particle emissions is needed when considering the potential health impacts of $LDSA^{al}$ exposure.

CRediT authorship contribution statement

Teemu Lepistö: Conceptualization, Methodology, Validation, Formal analysis, Investigation, Data curation, Visualization, Writing – original draft, Writing – review & editing. **Henna Lintusaari:** Conceptualization, Methodology, Investigation, Data curation, Visualization, Writing – review & editing. **Anna Oudin:** Conceptualization, Writing – review & editing. **Luis M.F. Barreira:** Investigation, Writing – review & editing. **Jarkko V. Niemi:** Resources, Writing – review & editing. **Panu Karjalainen:** Methodology, Investigation, Project administration, Funding acquisition, Writing – review & editing. **Laura Salo:** Investigation, Writing – review & editing. **Ville Silvonen:** Investigation, Writing – review & editing. **Lassi Markkula:** Investigation, Writing – review & editing. **Jussi Hoivala:** Investigation, Writing – review & editing. **Petteri Marjanen:** Investigation, Writing – review & editing. **Sampsa Martikainen:** Investigation, Writing – review & editing. **Minna Aurela:** Investigation, Writing – review & editing. **Felipe Reyes Reyes:** Resources, Writing – review & editing. **Pedro Oyola:** Resources, Writing – review & editing. **Heino Kuuluvainen:** Supervision, Writing – review & editing. **Hanna E. Manninen:** Resources, Writing – review & editing. **Roel P.F. Schins:** Resources, Project administration, Funding acquisition, Writing – review & editing. **Michal Vojtisek-Lom:** Methodology, Resources, Writing – review & editing. **Jakub Ondracek:** Resources, Writing – review & editing. **Jan Topinka:** Resources, Project administration, Funding acquisition, Writing – review & editing. **Hilkka Timonen:** Methodology, Project administration, Funding acquisition, Writing – review & editing. **Pasi Jalava:** Supervision, Project administration, Funding acquisition, Writing – review & editing. **Sanna Saarikoski:** Methodology, Investigation, Project administration, Funding acquisition, Writing – review & editing. **Topi Rönkkö:** Conceptualization, Methodology, Supervision, Project administration, Funding acquisition, Writing – review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

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