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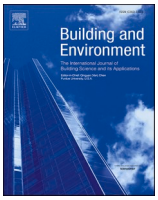

Lung-depositing surface area (LDSA) of particles in office spaces around Europe: Size distributions, I/O-ratios and infiltration

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ABSTRACT

Air pollution, and specifically particulate matter pollution, is one of the greatest dangers to human health. Outdoor air pollution ranks third in causes for premature death. Improving indoor air quality is of immense importance, as the time spent indoors is often much greater than the time spent outdoors. In this experimental study, we evaluate the levels of particle pollution in indoor air in four offices across Europe, compare the indoor particles to outdoor particles and assess where the particles originate from. The measurements were conducted with an Electrical Low-Pressure Impactor (ELPI+) for particles between 6 nm and 1 μm. The chosen metric, lung-deposited particle surface area (LDSA), targets the health impacts of particle pollution. Based on the measurements, we determined that most of the indoor air particles infiltrated from outdoor air, although two of the offices had very limited indoor activity during the measurement campaigns and may not represent typical use. The highest median indoor LDSA concentration during daytime hours was 27.2 $\mu m^2/cm^3$, whereas the lowest was 2.8 $\mu m^2/cm^3$. Indoor air in general had lower LDSA concentrations than outdoor air, the corresponding outdoor LDSA concentrations being 35.8 $\mu m^2/cm^3$ and 9.8 $\mu m^2/cm^3$. The particle size ranges which contributed to the highest concentrations were 50–100 nm and 300–500 nm. These size ranges correspond to soot mode and accumulation mode particles, which represent local and regional sources, respectively. Based on this study, limiting particle infiltration is the key factor in keeping indoor air in offices free of lung-depositing particles.

1. ıntroduction

Particles in ambient air originate from both natural sources (sea spray, wildfires, new particle formation from precursor compounds emitted by vegetation, etc.) and anthropogenic sources (traffic, wood combustion, industrial processes, etc.). An abundance of studies, both epidemiological [1] and toxicological [2], have shown that aerosol particles are detrimental to human health. Air pollution (including indoor and outdoor particles as well as ozone) is the third largest risk factor for death after smoking and high blood pressure [1], and unlike the first two risk factors, individuals have little to no control over their own exposure levels.

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In 2021, the World Health Organization (WHO) expanded their air quality guidelines [3] to include indoor air (previously, the guidelines were only applicable to outdoor air). This is an important update, as in many places the majority of time is spent indoors. For example, in a 2007 study [4], Helsinki residents spent 22.1 h/day indoors and Prague residents 22.7 h/day. In Germany, the average time spent indoors at home was found to be 15.7 h/day, not including time spent indoors at other locations [5]. Indoor aerosols can differ from outdoor aerosols due to many factors. First, indoor sources contribute much more to indoor air than to outdoor air, while outdoor sources contribute to both—depending on the type of ventilation. Ventilation also determines how quickly an emitted aerosol dilutes and is removed from the indoor space. Because of the low light intensities indoors [6], oxidants such as O_3, OH and NO_3 are produced at a decreased rate [7] and the aging of aerosol particles is much slower compared to outdoors, although new particle formation can happen, e.g., following the usage of cleaning products [8] or painting [9]. Finally, indoor spaces have much more surface area than outdoor spaces, allowing particle deposition and resuspension to play a larger role and increasing the significance of multiphase chemistry in indoor surfaces [10].

In addition to expanding the guidelines to cover indoor air, the updated WHO guidelines give a good practice statement regarding ultrafine particles (smaller than 100 nm), recommending particle number concentration measurement of particles with a lower limit of $\leq$ 10 nm. While no guideline limits were given, the document regards a 1-h average of 20 000 1/cm^3 and above as a high concentration, as well as a 24-h average of 10 000 1/cm^3 and above, whereas a low concentration is less than 1000 1/cm^3. These values help give context to measurements.

Epidemiologically, the harm caused by particles has been associated with the mass concentration of particles smaller than 2.5 μm ($PM_{2.5}$). However, ultrafine particles have been found to be more toxic than can be explained by their mass concentration [11] and toxicological studies suggest that particle surface area concentration is a better estimate of toxicity for insoluble particles [12,13]. Particle size is an important parameter because it determines where in the respiratory system the particle is likely to deposit [14]. Ultrafine particles also show an increased retention time in the lung, having a longer pulmonary clearance time than larger particles [15]. Furthermore, the chemical composition of particles can have a significant effect on health outcomes [16–18]. Ideally, all these metrics could be precisely and comprehensively measured. We must, however, choose metrics which are both relevant to health effects and practical to measure.

Measurements of lung-deposited surface area (LDSA) are gaining popularity, especially in indoor air [19–28]. LDSA refers to the surface area of particles, multiplied by their deposition probability in a specific region of the lung. In this study we consider only the deposition in the lung alveoli, which is the region where the interaction between the pulmonary circulation and the respiration occurs. Particles entering the lung alveoli may transport into other organs, the cardiovascular system [29] and the brain [30]. Thus, LDSA is a highly relevant metric in the study of potential harm to human health. Although evidence is still scarce due to a lack of data, an epidemiological study on household air pollution found LDSA exposure to be a superior metric to $PM_{2.5}$ for lung function [26]. Furthermore, previous studies indicate that LDSA correlates better with mortality [31] and subclinical atherosclerosis [32] than particulate mass. LDSA also has a higher sensitivity to local emissions, such as black carbon (BC, soot) compared to $PM_{2.5}$ [33,34]. Local emissions, especially traffic-related emissions, have been shown to have higher toxicity per PM mass than aged aerosol [35,36], but their contribution to total $PM_{2.5}$ mass is often not as significant. Thus, LDSA measurements are an important addition to complement the traditional $PM_{2.5}$ mass measurements.

In addition to the health-relevance, LDSA is practical to measure with electrical particles sensors which measure by charging particles using a corona wire or needle. The current measured from the charged particles can be converted to LDSA for particles within a range of ~20 nm–~300 nm [37,38]. These instruments are usually not as sensitive as, e.g., condensation particle counters (CPC), but a major benefit is their affordability and lower maintenance needs. The LDSA size distribution can be measured by an SMPS (Scanning Mobility Particle Sizer) by assuming spherical particles and calculating the surface area distribution based on the number size distribution, and then multiplying by the deposition curve. A similar method can be employed with other instruments which measure particle size distributions, such as the ELPI + [39,40]. Different parameterizations for particle lung deposition can be found in the literature, e.g. [41,42]; however, an often-cited empirical equation based on an ICRP model [43] is given by Hinds [14]. LDSA size distributions have great potential in health effect studies as they give hints of emission sources and composition of particles that are most relevant in terms of lung exposure.

The infiltration factor F_{inf} describes the infiltration of outdoor particles into indoor air. It corresponds to the fraction of outdoor particles that infiltrate into indoor air and remain suspended. As opposed to the indoor/outdoor ratio (the ratio of the indoor and outdoor particle concentrations), F_{inf} will only get values between 0 and 1 and it can be viewed as the ability of the building to resist the infiltration of outdoor particles into indoor air. The general definition for the infiltration factor using the indoor and outdoor particle concentrations C_{in} and C_{out} is

$$C_{in} = F_{inf} C_{out} + C_{\dot{S}}, \quad (1)$$

where $C_{\dot{S}}$ is the indoor concentration originating from indoor sources [44]. Linear regression can be used to easily find F_{inf} and $C_{\dot{S}}$, when C_{in} and C_{out} are measured.

Currently, infiltration of LDSA from outdoor to indoor air is poorly understood. A study on commercial buildings with mechanical ventilation reported size-segregated infiltration of particle mass as well as the number of ultrafine particles [45]. They found that infiltration was low for ultrafine particles (0.27), higher for particle mass of sizes 300 nm to 1 μm (>0.4) and lower again for particles up to 10 μm (<0.4). They also looked at black carbon infiltration, which was especially high at 0.54. These findings suggest that LDSA infiltration may also be high as it is highly sensitive to the particle size range which most easily penetrates into indoor air. However, we could not find previous studies of LDSA infiltration from outdoor to indoor air. Furthermore, while the common sensor measurement of LDSA is valuable for measuring in the size range it is targeted to, recent articles point out that accumulation mode particles (somewhat aged particles, originating from regional sources) also contribute to the total LDSA [34,46]; therefore, wider size range measurements, including size distributions, are necessary to understand the potential health impacts of outdoor air pollution in indoor air.

The aim of this study is to better understand how outdoor air pollution contributes to indoor air in terms of LDSA. Infiltration of LDSA is highly relevant because of its direct linkage to health and because the metric is sensitive to soot and accumulation mode particles i.e., to the most penetrating particle size range (~200 nm). We measured the LDSA particle size distribution from indoor and outdoor air in four offices located in Finland, Germany, and Czechia. Based on the measurements, we evaluated the role of particle infiltration from outdoor to indoor air as well as the contribution of indoor sources. The most likely particle emissions sources are discussed based on previous research on ambient particle size distributions. Understanding particle infiltration allows for more effective strategies to combat indoor particle pollution and minimize health risks. To the best of our knowledge, our paper is first to report LDSA infiltration factors and LDSA particle size distributions in offices.

2. METHODS

2.1. MEASUREMENT SITES AND CONDITIONS

To better understand the environments in which our study was conducted, we gathered general information as well as particle concentrations and other metrics for each of the four studied cities. These are presented in Table 1. The air quality data is from the measurement station nearest to each of the offices under study. The four offices and their related parameters are listed in Table 2. Local maps for each office are included in Figs. S1–4. These figures also show the average indoor and outdoor LDSA concentrations during different wind conditions.

Air quality in Tampere is very good: in 2020–2021 Tampere had one of the lowest $PM_{2.5}$ levels in Europe (in comparison to more than 300 cities). The office in Tampere is in an industrial area a few kilometers from the center of the city. Nearby is a train yard and highway, and the office is surrounded by industrial buildings and detached housing. The other three offices were located within their respective inner cities; thus, the main source of local emissions is most likely traffic.

In the center of Helsinki, traffic is an important source of particle pollution [47]. Our measurements were in early spring, when a common air pollution issue is episodes of high PM_{10} caused by studded tires grinding the road after the snow melts [48]. The Helsinki office was located along a narrow street with two traffic lanes (one in each direction), and near areas with marine traffic. The closest major harbors serving cruise and cargo ships are Länsisatama (1.5–2.0 km southwest), Katajanokka terminal (1 km east) and Olympia terminal (0.5 km southeast). A smaller harbor, Eteläsatama, serving ferries, is located only 250 m eastward. Heavy transport vehicles supply the Olympia terminal through a road located one block (100 m) away from the measurement site. The icebreakers fleet operates from a harbor 1.5 km eastward.

Despite the low annual average concentrations in Finland, in wintertime, a temperature inversion can form a layer of cold air near the surface trapped by the warmer air above, with any low-level emissions trapped as well [49,50]. This can occur under cold, low wind conditions. Often the cold temperature also leads to increased residential wood combustion in fireplaces and sauna stoves, resulting in episodes of increased pollution [51]. Additionally, long-range transport (LRT) episodes can occur in Finland several times a year, contributing especially to the number of accumulation mode particles [52]. During the measurements in Helsinki, an LRT-episode affected measured concentrations for some days. During a part of the Tampere measurement campaign, the particle concentrations were quite high, which was most likely due to a combination of temperature inversion and high emissions from biomass combustion for heating. These episodes are discussed in more detail in Section 3.

The air quality in Düsseldorf was within the limit values for the EU regulated parameters (including $PM_{2.5}$) in 2021. The mean $PM_{2.5}$ is similar to values observed for other German cities. The introduction of the low emission zones in many German cities (since 2009 in Düsseldorf) has led to a significant improvement in air quality, including particulate matter pollution. During the measurement campaign in Düsseldorf, the weather conditions were typical for the time of year. The road adjacent to the measurement site has an estimated traffic volume of ~20 thousand vehicles per day. In Düsseldorf, the office was in an urban area near the city center, along a street with two lanes in both directions and a tramline in between.

The $PM_{2.5}$ and PM_{10} levels in Prague are similar to Düsseldorf, as shown in Table 1. Coal as a source of heat in homes was largely abandoned in the 90's, significantly improving air quality [53]. However, as a consequence, road traffic has risen to the be the largest contributor to air pollution, especially of ultrafine particles [54]. Although engine exhaust emissions have been reduced by better technology, the number of vehicles has continued to increase and related pollution concentrations have leveled or even increased [53], representing a serious health problem. The Prague office was along a narrow street, with tram lines going in both directions. The intensity of traffic in this area was approximately 20 000 cars per day and about 400 trams a day in 2021.[3]

The office rooms in Helsinki and Prague were unused during the measurements except for when the rooms were visited by the researchers: in Helsinki this was due to the COVID-19 pandemic, and in Prague it was not possible to set up the measurements in a way that would have allowed the usage of the room. In Tampere, the room was used for 4 h a day on average as a meeting room, and the Düsseldorf office was a normal office with several people working normal hours.

Table 1
Air quality related statistics for each of the four studied cities.

	Tampere	Helsinki	Düsseldorf	Prague
City area (km^2)	523	214	217	496
Population (2021)	244 000	658 000	619 000	1 301 000
Registered passenger cars	98 000[a]	255 100[b]	360 000[c]	902 000[d]
$PM_{2.5}$ annual mean (2020–2021)[e]	4.1	5.2	10.2	12.2
PM_{10} annual mean (2021)[f]	11.1	19.6	19.3	22.6
Station code for PM_{10} site	FI00549	FI00564	DENW082	CZ0ALEG

[a] City of Tampere, (Transport, streets and maintenance), 2023. Retrieved August 1, 2023, from: https://www.tampere.fi/liikenne-kadut-ja-kunnossapito/liikennemaarat.

[b] City of Helsinki, 2023. Retrieved August 1, 2023, from: https://www.hel.fi/helsinki/fi/kartat-ja-liikenne/kadut-ja-liikennesuunnittelu/tutkimus-ja-tilastot/moottoriajoneuvoliikenteen-maarat/

[c] State Office Nordhein-Westfalen, (Statistics and IT services), 2023. Retrieved August 1, 2023, from: https://www.it.nrw/statistik/wirtschaft-und-umwelt/verkehr.

[d] Prague Transportation Yearbook, 2020. Retrieved August 1, 2023, from: https://www.tsk-praha.cz/static/udi-rocenka-2020-vm-cz-HTML/index.html

[e] European Environment Agency, 2023. Retrieved August 1, 2023, from: https://www.eea.europa.eu/themes/air/urban-air-quality/european-city-air-quality-viewer

[f] European Environment Agency, 2023. Retrieved August 1, 2023, from: https://discomap.eea.europa.eu/map/fme/AirQualityExport.htm

2.2. MEASUREMENTS AND INSTRUMENTATION

Measurements were conducted in 2021 and 2022, with each of the measurement campaigns lasting 2–3 weeks. The main particle instrument was the ELPI + by Dekati Ltd. In Tampere and Helsinki, we used two separate instruments, one for outdoor and one for indoor air, alternating the indoor measurement between room air and supply air from the mechanical ventilation with an automatic three-way valve set to 15-min intervals. In Tampere, the mechanical ventilation was switched off during nighttime beginning on the evening of December 13th as another part of the project required analysis of the indoor environment during and after the change to the ventilation. In Prague and Düsseldorf there was no mechanically provided supply air, and only one instrument was used for the measurements, alternating between outdoor and indoor air, again with 15-min intervals. In the following sections, the offices are referred to by their respective cities.

The indoor sampling point in each location was at least 1 m away from the walls, and at a height of 1.5 m. In Tampere, Düsseldorf and Prague, the outdoor sample was taken just outside the office window and in Helsinki on the roof of the building, just outside the ventilation fresh air intake. The fresh air intake was on the courtyard side of the building, at ~21 m above ground and the office was facing the road, at ~12 m above ground. In Tampere, the office was located on the ground

[3] Technická správa komunikací hlavního města Prahy. Retrieved August 1, 2023, from: https://www.tsk-praha.cz/wps/portal/root/dopravni-inzenyrstvi/intenzity-dopravy.

Table 2
Parameters of the studied offices and locations.

	Tampere	Helsinki	Düsseldorf	Prague
Building year	1971	1972	1969	1861
Ventilation type	Mechanical	Mechanical, partially recirculated	Exhaust-only	Natural
Filter grade	G5	F7	–	–
Supply air flow (m^3/h)	313	331	–	–
Room volume (m^3)	124	66	52	100
Room floor area (m^2)	43	25	17	20
Estimated room usage (%)	50	10	90	10
Outdoor temperature mean (min … max) (°C)	−6.5 (−22.8 … 3.5)[a]	0.9 (−8.2 … 8.6)[a]	9.3 (−1.0 … 19.2)[b]	6.4 (−1.4 … 9.3)[c]
Measurement period	30.11.2021–20.12.2021	12.3.2021–29.3.2021	8.3.2022–23.3.2022	25.3.2022–4.4.2022

[a] Finnish Meteorological Institute open data, Weather observations. Retrieved August 1, 2023, from https://en.ilmatieteenlaitos.fi/download-observations.
[b] Deutscher Wetterdienst. Retrieved August 1, 2023, from https://www.dwd.de/EN/ourservices/cdc/cdc_ueberblick-klimadaten_en.html.
[c] Visual Crossing Corporation. (2023). Visual Crossing Weather (2022). [data service]. Retrieved August 1, 2023, from https://www.visualcrossing.com/.

floor and the measurement instruments were in the ATMo-Lab mobile laboratory vehicle [55] parked next to the building. The indoor sample was drawn to the instruments through the ventilation window with a heated sample line kept between 10 and 30 ° C to minimize possible condensation effects. The window was fitted with a heat insulating panel that had an opening for the sample line, and the panel and opening were sealed with tape. The outdoor sample was taken 2 m away from the wall at a height of 1.5 m. In Düsseldorf, the office was located on the 1st floor counting up from ground level. An opening was made through the window frame, and the sample line was fitted through the opening that was sealed with tape. The sampling point was ~5.5 m above ground level and 0.3 m away from the wall. The horizontal distance to the road was ~13 m. In Prague, the office was also located on the 1st floor and the sampling point was at the same height and distance from the wall as in Düsseldorf. It was not possible to make an opening in this building, and the window was kept slightly open to fit the sample line outside. The gaps were sealed with tape. The horizontal distance to the road was ~3 m.

The two-to-three-week measurement campaigns in each office were mostly successful. In Prague and Helsinki, there are some gaps in the data due to problems with the instruments. In the time series data plots, all recorded data is presented, whereas plots with time-averaged data only present the periods where both indoor and outdoor data was recorded to allow for comparison between indoor and outdoor air.

The ELPI+ (abbreviated from Electrical Low-Pressure Impactor) is a cascade impactor where charged particles are measured electrically from each impactor stage [39]. After fourteen impactor stages (one pre-impactor and thirteen measured stages) the very smallest particles are gathered with a filter, also measured electrically. The smallest measurable size is approximately 6 nm, limited by the particle charging efficiency. The largest measured size is 10 μm; however, in this study we have limited our analysis to sub 1 μm particles. The measured currents are first corrected to a known zero level (achieved by periodically measuring clean air through a HEPA-filter), then currents are adjusted for sub-cut deposition [56]. Finally, the LDSA concentration of each impactor stage is calculated by multiplying the current with a stage-specific conversion factor [40]. The conversion factors are based on the equations given by Hinds [14], following the ICRP model [43]. Five different ELPI + units were used during the measurements. The locations where each unit was used are presented in Table S1. Each unit is calibrated by the manufacturer and this calibration is used in the conversion from the measured electrical current to LDSA.

The lung deposition is affected by the hygroscopicity and shape of the particles; however, incorporating these factors to the model would require extensive additional measurements. A previous study has shown that particles from both wood combustion and traffic exhaust tend to have low hygroscopicity—growth factors lower than 1.33 [57]. On the other hand, larger background particles tend to be hygroscopic, which could increase their alveolar deposition [57]. Another uncertainty related to lung deposition is its human component, as lung morphology and breathing patterns vary from person to person. In offices, the breathing patterns between individuals can however be assumed to have less variation than in other occupational settings, where physical activity such as walking or heavy lifting are more prevalent. In the equations given by Hinds [14], the deposition efficiency is an average of variable breathing rates and different genders.

ELPI + categorizes particles based on their aerodynamic diameter. Low-density particles will therefore be recorded as being smaller than their geometric size, increasing the surface area contribution of small particles while decreasing the contribution of large particles. On the other end, dense particles will be recorded as larger than their geometric particle size. From the lung-deposition viewpoint, both the geometric size and the aerodynamic size are important. Diffusional movement is governed by the geometric size, whereas inertial impaction depends upon the aerodynamic size. As particle density was not measured, we conducted the data analysis with the assumption of spherical particles with unit density.

The original 1 Hz data is presented as 30-min averages — both to remove noise and to have corresponding indoor and outdoor data points for each averaging period (as the automatic valve was controlled to switch every 15 min). From the averaged data, those data points having a coverage of less than 25 % were removed. Thus, each 30-min averaged data point includes data from at least 7.5 min of sampling.

3. Results & discussion

3.1. Temporal variations of LDSA concentrations

Fig. 1 shows the total measured LDSA concentration for indoor, outdoor and supply air (where applicable). Note that the y-axis is logarithmic, as concentrations varied highly. The indoor air LDSA concentrations were almost always lower than the outdoor concentrations, bar a few peaks. This points to the outdoor air being the main source of LDSA indoors. In Helsinki, the supply air concentration was slightly lower than indoor concentration, whereas the opposite was true in Tampere.

In Tampere and Helsinki, the mean LDSA concentrations during the measurement period were 16.3 and 10.7 $\mu m^2/cm^3$ outdoors and 6.3 and 2.6 $\mu m^2/cm^3$ indoors, respectively, whereas in Düsseldorf and Prague the mean concentrations were 35.1 and 30.8 $\mu m^2/cm^3$ outdoors and 18.8 and 21.2 $\mu m^2/cm^3$ indoors, respectively. Here we only considered the time periods where both indoor and outdoor data was available. Table S2 also lists the median LDSA concentrations and the standard deviations. The mean indoor/outdoor ratio of LDSA was highest in Prague (0.77) and lowest in Helsinki (0.31), with Tampere (0.44) and Düsseldorf (0.59) in between. The median I/O ratios and standard deviations are listed in Table S2. The daily minimum concentrations, which can be considered as indications of background concentrations in the absence of local sources, were estimated as the 1st percentile of the daily 1 Hz measurement data. In Tampere and Helsinki, the mean daily

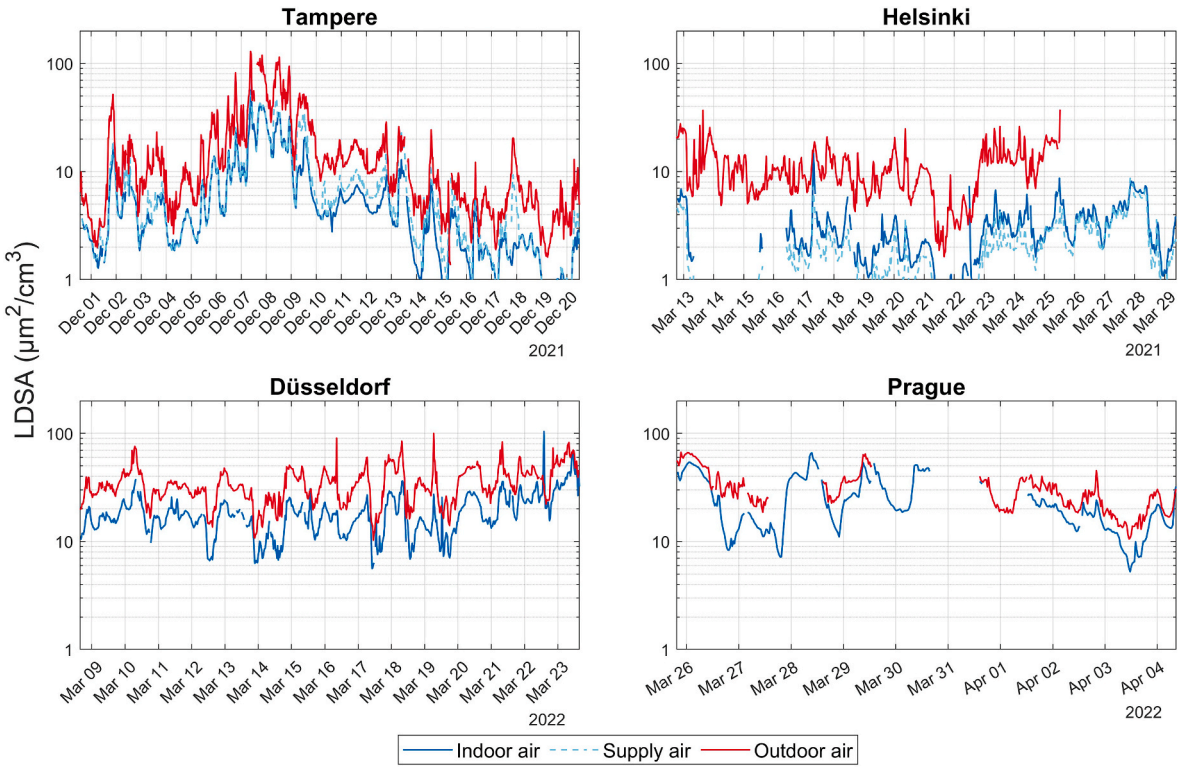

Fig. 1. Total LDSA concentration timeseries at each location for indoor, outdoor and supply air (where applicable). Note the logarithmic y-axis.

minimum LDSA concentrations were 6.0 and 6.2 $\mu m^2/cm^3$ outdoors, and 2.8 and 1.2 $\mu m^2/cm^3$ indoors, respectively, whereas in Düsseldorf and Prague the outdoor mean daily minimum concentrations were 17.0 and 15.5 $\mu m^2/cm^3$ corresponding to 9.7 and 8.6 $\mu m^2/cm^3$ indoors, respectively. Variations in the daily minimum concentrations are shown in Table S3.

Fig. 2 shows the LDSA distributions as a time series both outdoors and indoors at each measurement site. The median particle size of the distribution is depicted with a black line. The plots show that there are faster changes in the concentration of smaller particles (under 100 nm), than larger particles (roughly 200 nm–700 nm), indicating that the smaller particles are likely to be from local sources. Comparing the indoor and outdoor time series shows that the indoor concentrations were smaller than the corresponding outdoor concentrations. In Helsinki, Düsseldorf and Prague, the concentrations follow a clear diurnal pattern, better seen in Fig. S5, where a rise in concentration is visible in the morning, likely to be from local traffic. In Helsinki, the highest concentration is visible at 8:00–9:00, in Düsseldorf at 6:00–7:00 and in Prague at 9:00–10:00, due to the different rhythm of traffic density in the cities. Interestingly, the lowest concentrations in Düsseldorf occur in the late afternoon. Histogram plots of the median particle size are included in Fig. S6.

In the Helsinki LDSA timeseries, we identified a long-range transport episode beginning on March 24th, characterized by the sudden appearance of an accumulation mode, which is especially prominent in the outdoor LDSA time series. We confirmed the regional nature of the new mode by observing a simultaneous increase in $PM_{2.5}$ at an urban background station, seen in Fig. S7, and by comparing the air mass back trajectories before and during the event, seen in Fig. S8. In Tampere, an atmospheric inversion occurred between December 7th and December 9th, confirmed by low wind speeds, low temperatures and a low boundary layer height seen in Fig. S9. The influence of these episodes on particle infiltration and indoor LDSA concentrations is discussed further in relation to Fig. 4.

Several previous studies report LDSA concentrations from indoor air, although most studies are from residential buildings, where cooking plays an important role in concentrations [25]. A study on air quality in Korean daycares reports mean LDSA from 16.3 $\mu m^2/cm^3$ to 78.8 $\mu m^2/cm^3$ [27]. Canha et al. [21] found that LDSA ranged from 7.3 to 95.2 $\mu m^2/cm^3$ in bedrooms in Portugal during nighttime. These ranges are similar to our indoor air results. The outdoor concentrations can be compared to previous studies of outdoor LDSA. Previous measurements in Helsinki found the mean LDSA in a street canyon to be 22 $\mu m^2/cm^3$ over a one-year measurement [58]. A year-long study conducted in London found that quieter areas of the city had LDSA concentrations around 15 $\mu m^2/cm^3$, while areas with major roads and high numbers of restaurants had concentrations around 25 $\mu m^2/cm^3$ [59]. The mean LDSA outdoors in our study ranged from 11 $\mu m^2/cm^3$ in Helsinki to 35 $\mu m^2/cm^3$ in Düsseldorf, which again is within a similar range to previous studies.

3.1.1. *Size distributions of LDSA and I/O ratio*

Fig. 3 shows the median LDSA particle size distributions for room air, supply air (when available) and outdoor air. On the right-side y-axis is the indoor to outdoor LDSA ratio as a distribution. Tampere, Helsinki, and Düsseldorf have approximately bimodal LDSA distributions, while Prague has a separate third mode in the nucleation mode size range. With a finer measurement of particle size, perhaps also the Düsseldorf distribution might display a separate nucleation mode. The outdoor distribution from Helsinki also displays many particles in the sub-20 nm size range. The particle size ranges which contribute most to the indoor LDSA concentration in all studied offices are 50–100 nm and 300–500 nm. These size ranges correspond to soot mode and accumulation mode particles, which represent local and regional sources, respectively. LDSA particle size distributions with a linear y-axis are presented in Fig. S10.

In Finland, a bimodal distribution is typical when there is a local source of pollution from traffic and a long-range transport event which brings in larger particles [34,60]. However, wood combustion emissions are typical in a similar size-range to long-range transport aerosol [61,62]. The measurements in Helsinki were conducted in spring, and there

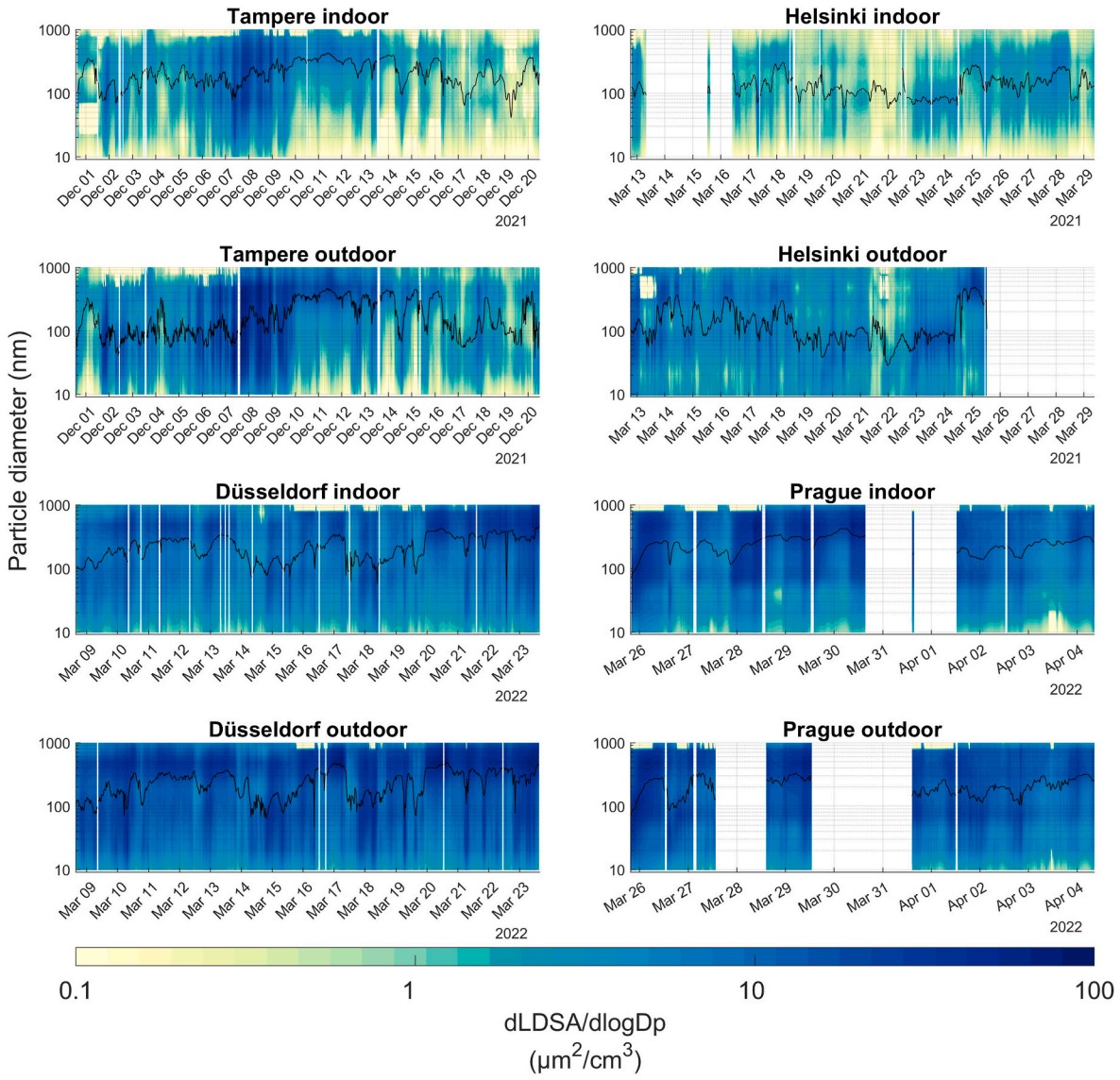

Fig. 2. The outdoor and indoor LDSA distributions over time at each location as well as the median particle size (black line). The data has been averaged into 30-min sections.

are very few houses in the area with wood fireplaces or stoves, thus an LRT episode is a far more likely explanation for the second mode. The size distribution during the LRT episode is shown in Fig. S11, displaying highly elevated accumulation mode particle concentrations typical of an LRT episode. In Tampere, on the other hand, the measurements took place mid-winter and adjacent to a suburban area with detached housing, where wood burning is common for heating in the wintertime. During an atmospheric temperature inversion episode on December 7th – 9th, local emissions were trapped near the ground resulting in high concentrations. Later, temperatures warmed up and the concentrations were lower. This also explains why the data from Tampere has such a large variance. Size distributions during the inversion event and on other days are compared in Fig. S12.

In Central European countries, a particle mode at particle sizes of a few hundred nanometers is typically a consistent contributor to LDSA [60]. This mode is also quite consistently present in the Prague and Düsseldorf time series shown here, in particle sizes 300–700 nm. This is within the accumulation particle size range, inferring a consistent presence of aged regional particles. Unlike in Finland (and other northern European countries) it is not as easily cleared by winds, as cities are located close together, and wind from any direction will bring a new batch of aged emissions. In Düsseldorf the concentration of accumulation mode particles follows a diurnal cycle, with a minimum concentration at 16:00 (Fig. S5), following the diurnal cycle of the boundary layer height (BLH). The diurnal cycle of the BLH and wind speed in each city can be seen in Fig. S13.

Indoor/outdoor LDSA ratios were below one in all four cases, and there was a clear correlation between the indoor and outdoor concentrations, indicating that the indoor concentrations were governed by the outdoor concentrations. This is in line with previous studies looking into indoor/outdoor particle concentrations in offices [63]. The size distribution of the I/O ratio has a peak near 200 nm in all the studied offices. This is most likely due to the size-dependency of the penetration efficiency of particles through the building envelope and supply air filters. Although the total penetration may vary significantly between buildings, the size-dependency of penetration typically shows a similar trend, the most penetrating particle size being between 100 nm and 300 nm [64,65]. Additionally, the deposition loss rate of particles to indoor surfaces is at its lowest for particles of this size [65]. In Tampere and Düsseldorf, the I/O ratio rises notably for particles around 750 nm. Based on previous research, particle penetration typically decreases for particles over ~500 nm [44,66,67]. This indicates that indoor sources

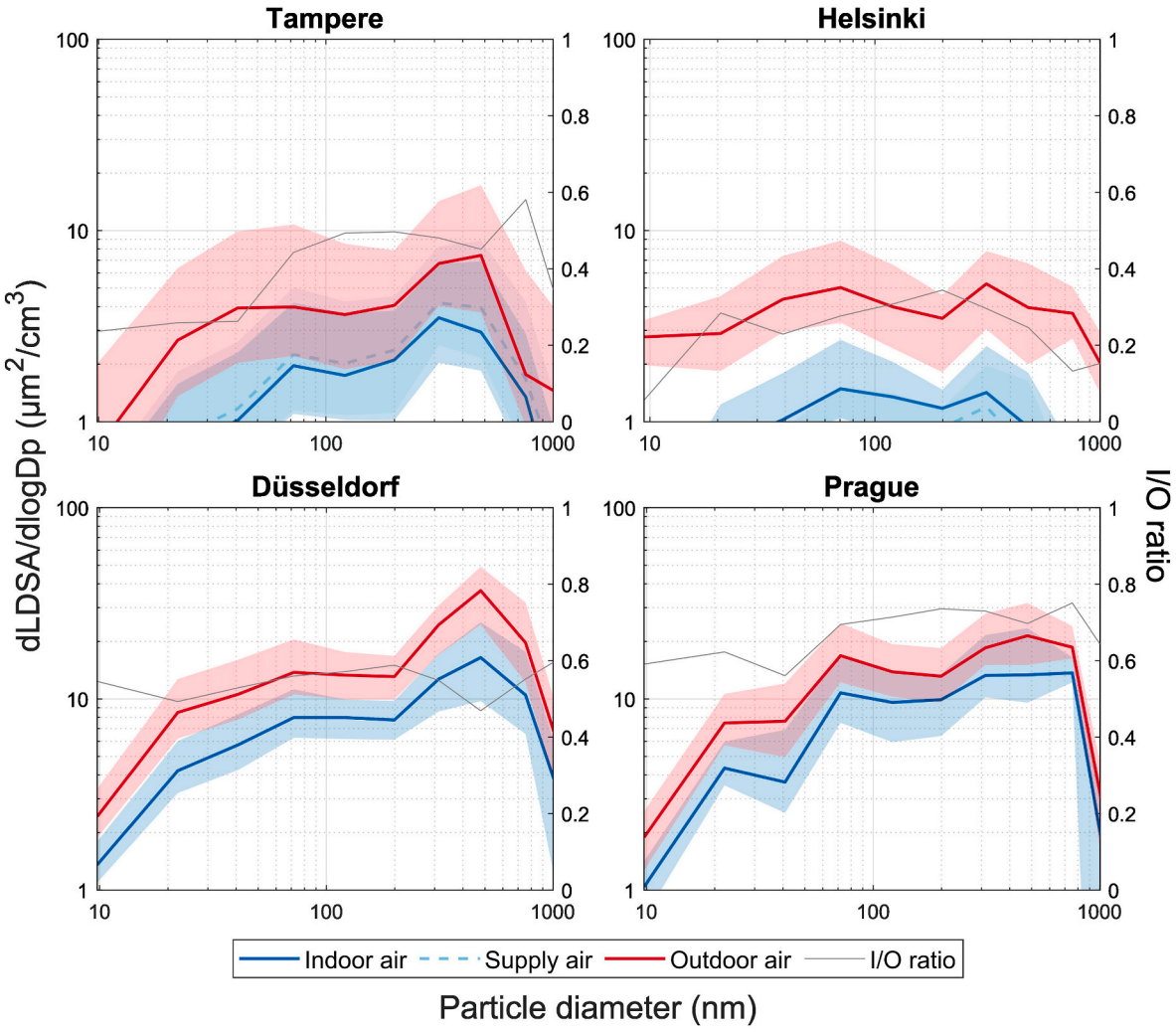

Fig. 3. Median LDSA particle size distributions and indoor-outdoor ratio for each office. The shaded area shows the 25th and 75th percentiles. Only data for which both outdoor and indoor distributions (or supply if applicable) were available are shown.

contributed to indoor particles around the 750 nm size. Most likely they are particles resuspended to the air due to people moving around in the room, as no other significant indoor particle sources were identified. It is however possible that they originate from another indoor source that we failed to identify; particles have been shown to transport between indoor environments [68,69]. In all the studied locations, an increase or leveling-off of the I/O ratio can also be seen for particles around 20 nm in size. A possible explanation for this is the formation of secondary organic aerosol (SOA) from gaseous indoor precursors [70,71]. The interquartile ranges of the I/O ratios are presented in Fig. S14.

3.1.2. *Particle infiltration*

Fig. 4 shows the correlation between indoor and outdoor LDSA concentrations. For each data set, a best fit line according to equation (1) is shown along with the line equation, where the infiltration factor F_{inf} is the slope and the contribution from indoor sources C_s is the y-intercept. All data points are shown with a grey dot and data from working hours is also circled. This was done especially since indoor sources may have a higher contribution during working hours, although from the results it is apparent that the indoor contributions were small even during working hours.

The offices using mechanical ventilation with supply air filters (Tampere and Helsinki) have significantly lower infiltration factors than the other two offices. The highest infiltration is observed in Prague, where no mechanical ventilation was utilized. In Dusseldorf only the exhaust air flow was mechanically controlled. Based on the linear fits, the contribution of indoor sources to the total indoor concentration was minor in all the studied locations. The strongest correlation between outdoor and indoor air was in Tampere, where the supply air filter efficiency was significantly lower than in Helsinki (20 % for 200 nm particles compared to 50 % in Helsinki). Helsinki had the weakest correlation, which can be related to the use of supply air recirculation; particles produced in other parts of the building may be transported to the studied room, and these would not be measured by the outdoor sampling. Another notable factor is the sampling point of outdoor air; it was sampled from the roof of the building, whereas in the other offices the outdoor sample was taken next to the office room window. The efficient supply air filter may also contribute to the weaker correlation. Finally, any large deviations from the general concentration trends will influence the correlation coefficient. These deviations are more common when using a relatively short averaging period. The linear fits in Fig. 4 are calculated using ordinary least squares fitting, where outlier data points have a large influence on the fit. Using a robust linear fitting method, the correlation coefficient R^2 between indoor and outdoor concentrations was notably higher in Helsinki and Düsseldorf (0.76 and 0.87 respectively, when considering all hours), where some clear outlier data points can be observed. The robust linear fits are presented in Fig. S15. Linear fits with 12-h averaged data are presented Fig. S16, where higher correlation coefficients are also observed.

Table 3 lists the mean and median indoor and outdoor LDSA

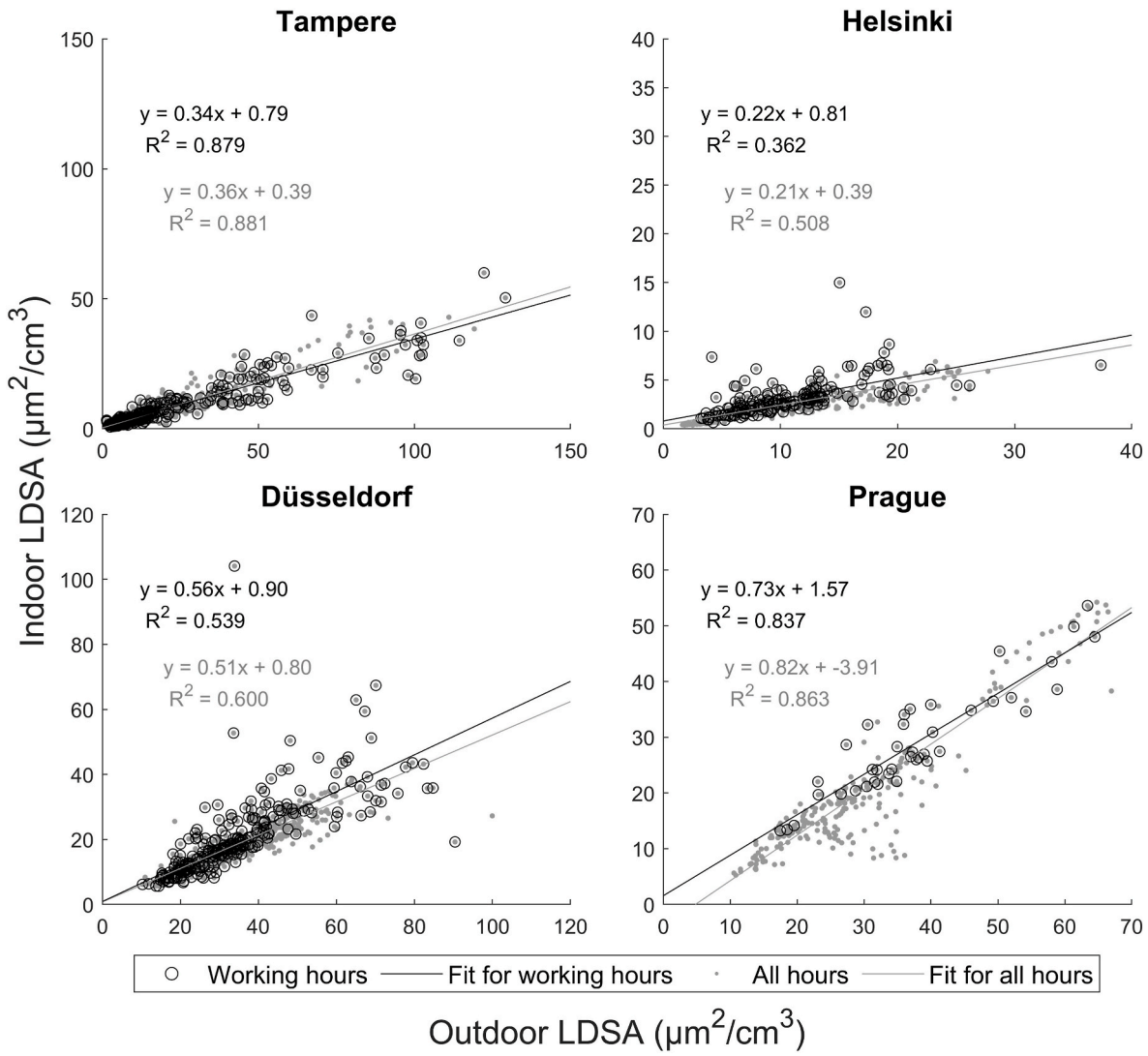

Fig. 4. Indoor versus outdoor LDSA scatter plots with fitted lines. Data from all hours is shown with grey dots and line. Working hours data, shown as black circles and line, is from weekday daytime hours (6:00 to 18:00).

Table 3
The median and mean indoor and outdoor LDSA concentration for working hours (weekdays from 6:00 to 18:00), along with standard deviation (STD) were calculated from the original 1-Hz data. The mean and median I/O ratio for LDSA concentration (6–1000 nm) along with standard deviation, the infiltration factor (F_{inf}), LDSA concentration contributed from indoor sources (C_s) and the correlation coefficient for determining the infiltration factor (R^2) were calculated from the 30-min averaged data. Data which was missing from either indoor or outdoor concentrations (within a period of 30 min) has been excluded.

		Tampere	Helsinki	Düsseldorf	Prague
Indoor	**Median ($\mu m^2/cm^3$)**	4.12	2.82	17.50	27.17
	Mean ($\mu m^2/cm^3$)	8.15	3.18	20.48	29.29
	STD ($\mu m^2/cm^3$)	9.17	1.94	12.97	9.58
Outdoor	**Median ($\mu m^2/cm^3$)**	11.13	9.80	31.68	35.84
	Mean ($\mu m^2/cm^3$)	22.07	10.72	34.79	38.16
	STD ($\mu m^2/cm^3$)	26.36	5.32	33.39	14.05
I/O ratio	**Median**	0.39	0.23	0.52	0.69
	Mean	0.41	0.25	0.54	0.67
	STD	0.17	0.12	0.15	0.14
Infiltration	F_{inf}	0.34	0.22	0.56	0.73
	C_s **($\mu m^2/cm^3$)**	0.79	0.81	0.90	1.57
	R^2	0.88	0.36	0.54	0.84

concentrations in each office, as well as the calculated infiltration factors, concentrations from indoor sources and correlation coefficients. The relevance of the varying infiltration characteristics of the buildings is evident when comparing the median indoor and outdoor concentrations. Even though the median outdoor concentrations in Tampere and Helsinki are similar, there is a significant difference between them in the indoor concentrations. The effect of varying infiltration characteristics is even more visible when comparing Düsseldorf and Prague.

Infiltration plots for the inversion event in Tampere and the LRT episode in Helsinki are presented in Fig. S17. The infiltration factor in Tampere did not change significantly during the inversion event (0.31 for non-event, 0.29 for event), whereas in Helsinki the LRT episode resulted in a notable reduction in the infiltration factor (0.22 for non-episode, 0.17 for episode). The reason for this is evident from the size distributions in Fig. S11 and Fig. S12; the normalized size distribution in Tampere remained relatively stable throughout the event, but in Helsinki the sudden increase in the concentration of particles over 400 nm leads to reduced infiltration, due to their weaker penetration through the supply air filter and building envelope.

3.1.3. Limitations and implications

In general, data from indoor air is only applicable to the surroundings where the data was collected. This is true for these measurements as

well, especially for absolute concentrations. The composition of indoor air varies greatly, due to the variations in outdoor air but also because of human activity in the indoor setting. Buildings with similar technical characteristics can thus display a wide variability in the indoor air. It is unfortunate that two of the offices were not in normal use during the study; however, even in the offices which were being used, the contribution of indoor sources was small, as discussed earlier. The measurements in each office were done in one season during two-to-three-week periods. As can be seen from Fig. S11 and Fig. S17, the environmental conditions such as the outdoor particle size distribution may have a significant effect on infiltration, and determining a single infiltration factor for a building can be problematic.

Based upon the results of this study, LDSA concentration in offices can best be controlled by limiting particle infiltration. This can be done, for example, by improving filtration of the supply air in mechanically ventilated buildings or using air cleaners which circulate air through a filter in naturally ventilated rooms. Although the measured LDSA concentrations were not very high, reductions should still be made whenever feasible — good occupational hygiene fosters health and well-being of the employees as well as lowers illness-related costs.

Even if the outdoor concentrations in all the studied locations were similar, the difference between the indoor concentrations could be as high as 200–300 %, depending on the infiltration characteristics of the building. This implies a significant source of uncertainty for exposure-response models between different locations and could help in explaining the discrepancy in mortality rate between countries [1,72]. Ideally, all indoor spaces should have as low particle concentrations as possible. In the case of offices, considering the findings in this and other studies, outdoor air is the predominant source of indoor particle pollution. Renewing existing ventilation infrastructure or building completely new central ventilation systems may not be feasible ways to reduce the infiltration of outdoor air, but interventions such as portable air cleaner units [24] can easily be implemented in high-pollution environments. However, air cleaners can unintentionally act as sources of volatile organic compounds [73], and should thus not be implemented without considering possible side effects. The most effective way to reduce exposure to particulate matter is to prevent the emission of particles at the source.

These results offer insight into possible LDSA concentration ranges and contributing particle sizes in offices. We have included information regarding the buildings, office rooms and surroundings, enabling extrapolation of the findings. As LDSA is a particularly health-relevant metric, and typically corresponds well to the most penetrating particle size, it can be a useful tool in developing our understanding of the total human exposure to particulate matter pollution. Further studies are however needed to rigorously establish typical LDSA concentrations and infiltration factors in offices.

4. Conclusions

This study evaluated LDSA concentrations, particle size distributions and infiltration factors for four offices in four different cities in Europe. The mean LDSA concentrations indoors during work hours were lower than the corresponding outdoor concentrations, ranging from 3.18 $\mu m^2/cm^3$ in Helsinki to 29.29 $\mu m^2/cm^3$ in Prague.

Contributions to LDSA were mostly from outdoor particles infiltrating into indoor air. Indoor sources of LDSA were low in the studied offices. The infiltration factors during work hours were lower for Helsinki and Tampere (0.22 and 0.34, respectively), where the supply air was filtered, whereas infiltration factors were higher for Düsseldorf and Prague (0.56 and 0.73, respectively), where supply air was unfiltered.

This study showed that indoor air particles cover a large size range when measured as LDSA. Bi-modal LDSA distributions were present in all four cities included in this paper, and in Tampere, Düsseldorf and Prague accumulation mode particles formed the larger mode for most of the measurement duration.

CRediT authorship contribution statement

Ville Silvonen: Writing – review & editing, Writing – original draft, Visualization, Methodology, Investigation, Data curation, Conceptualization. **Laura Salo:** Writing – review & editing, Writing – original draft, Visualization, Project administration, Methodology, Investigation, Conceptualization. **Tuomas Raunima:** Resources, Investigation. **Michal Vojtisek-Lom:** Writing – review & editing, Resources, Investigation. **Jakub Ondracek:** Writing – review & editing, Resources, Investigation. **Jan Topinka:** Resources, Project administration, Funding acquisition. **Roel P.F. Schins:** Writing – review & editing, Resources, Project administration. **Teemu Lepistö:** Writing – review & editing, Investigation. **Henna Lintusaari:** Writing – review & editing, Investigation. **Sanna Saarikoski:** Supervision, Funding acquisition. **Luis Barreira:** Resources, Investigation. **Jussi Hoivala:** Investigation. **Lassi Markkula:** Investigation. **Ilpo Kulmala:** Investigation. **Juha Vinha:** Resources, Project administration, Funding acquisition. **Panu Karjalainen:** Writing – review & editing, Funding acquisition. **Topi Rönkkö:** Writing – review & editing, Supervision, Resources, Project administration, Methodology, Funding acquisition, Conceptualization.

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

Supplementary data to this article can be found online at https://doi.org/10.1016/j.buildenv.2023.110999.

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