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Personal exposure to gaseous and particulate phase polycyclic aromatic hydrocarbons (PAHs) and nanoparticles and lung deposited surface area (LDSA) for soot among Norwegian chimney sweepers

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ABSTRACT

Exposure to polycyclic aromatic hydrocarbons (PAHs) of high molecular weight from chimney soot can cause cancer among chimney sweepers. These sweepers may also be exposed to high concentrations of nanosized particles, which can cause significant inflammatory responses due to their relatively greater surface area per mass. In this study, the authors aimed to assess the exposure profiles of airborne personal exposure to gaseous and particulate PAHs, and real-time samples of the particle number concentrations (PNCs), particle sizes, and lung-deposited surface areas (LDSAs), for chimney sweepers in Norway. Additionally, the authors aimed to assess the task-based exposure concentrations of PNCs, sizes, and LDSAs while working on different tasks. The results are based on personal samples of particulate PAHs (n = 68), gaseous PAHs (n = 28), and real-time nanoparticles (n = 8) collected from 17 chimney sweepers. Samples were collected during a "typical work week" of chimney sweeping and fire safety inspections, then during a "massive soot" week, where larger sweeping missions took place. Significantly higher PAH concentrations were measured during the "massive soot" week compared to the "typical work week," however, the timeweighted average (TWA) (8-hr) of all gaseous and particulate PAHs ranged from 0.52 to 4.47 $\mu g/m^3$ and 0.49 to 2.50 $\mu g/m^3,$ respectively, well below the Norwegian occupational exposure limit (OEL) of $40 \,\mu g/m^3$. The PNCs were high during certain activities, such as emptying the vacuum cleaner. Additionally, during 2 days of sweeping in a waste sorting facility, the TWAs of the PNCs were 3.6×10^4 and 7.1×10^4 particles/ cm³ on the first and second days, respectively, which were near and above the proposed nano reference limit TWA value of 4.0×10^4 particles/cm³ proposed by the International Workshop on Nano Reference Values. The corresponding TWAs of the LDSAs were 49.5 and 54.5 μ m²/cm³, respectively. The chimney sweepers seemed aware of the potential health risks associated with exposure, and suitable personal protective equipment was used. However, the PNCs reported for the activities show that when the activities change or increase, the PNCs' TWAs can become unacceptably high.

Introduction

Approximately 900 chimney sweepers work in Norway today. In keeping with a tradition going back hundreds of years, they manually sweep chimneys with long steel brushes (IARC 2012). As such, chimney sweepers are exposed to polycyclic aromatic hydrocarbons (PAHs) of high molecular weight found in chimney soot during their work. At room temperature, these PAHs exist in both vapor and aerosol forms. The lipophilic properties of PAHs ensure that PAHs are easily absorbed in the epithelia of the respiratory tract and by the skin. The International Agency for Research on Cancer (IARC) has reviewed the cancer evidence among chimney sweepers. As a result, soot was classified as a Group 1 carcinogen for humans (IARC 2012). Soot is black particulate matter formed as a by-product of

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KEYWORDS

Cancer; DiSCmini; occupational exposure; personal protective equipment; sweeping; ultrafine particles

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combustion or pyrolysis of organic materials, including coal, wood, waste oil, paper, plastics, and other household refuse. The chemical constituents of soot vary considerably, depending on the origin fuel source. Generally, soot consists of up to 60% carbon, a high range of inorganic materials, and a soluble organic fraction. The inorganic fraction consists of different metals, salts, oxides, sulfur, and nitrogen compounds, while the PAHs are found in the soluble organic fraction (IARC 2012). The PAHs consist of more than 100 chemicals with at least two fused benzene rings (Santos et al. 2019). The PAH benzo[A]pyrene (BaP) is a Group 1 human carcinogen. Additionally, some other PAHs are classified as Group 2 A/2B, which include probable or possible human carcinogens (The Norwegian Labor Inspection Authority 2010).

In a recent analysis of 14 pooled case–control studies, chimney sweeping was one of the male occupations with the most frequent PAH exposure (Olsson et al. 2022). In a follow-up study spanning 1960 to 2005, which examined cancer incidence data by occupational category for 15 million people in five Nordic countries, chimney sweeping was among the occupations with the highest standardized incidence ratios (SIRs) when looking at all cancers combined. Examining the results from Norway alone (n=798), chimney sweepers had a significantly higher SIR of lung cancer (SIR = 1.77), as was the case in Denmark (SIR = 1.58), and Sweden (SIR = 1.71) (Pukkala et al. 2009).

In Norway, an 8-hr occupational exposure limit (OEL) for air exposure to the sum of 21 particulate PAHs has been set $(40 \,\mu g/m^3)$. Individual limit values also exist for two PAHs, namely, naphthalene $(50,000 \,\mu\text{g/m}^3)$, and biphenyl $(1,000 \,\mu\text{g/m}^3)$, however, no limit value for BaP exists (The Norwegian Labor Inspection Authority 2010). In the Netherlands, based on estimated excess cancer mortality rates due to 40 years of occupational exposure to BaP (4 per 1,000 and 4 per 100,000), tolerable and acceptable limit values of 550 and 5.7 ng BaP/m³ have been proposed, respectively (DECOS 2006). The Swedish Work Environment Authority (SWEA) has set an 8-hr OEL value for BaP exposure in the air at $2 \mu g/m^3$. For short-term BaP exposure, SWEA has set a maximum allowable concentration of 15 min to $20 \,\mu g/m^3$ (Swedish Work Environment Authority 2018).

Limited data on air exposure to PAH among chimney sweepers exist. In a study published in 1989, 20 different PAHs were analyzed across 115 samples, and the air concentrations of BaP were found to be on average 0.36, 0.83, and $0.82 \,\mu\text{g/m}^3$ for oil firing, oil solid firing, and pure solid fuel firing, respectively (Knecht et al. 1989). In a 1987 Swedish report, where exposures to total dust and PAHs were assessed, averages of $3-19 \,\text{mg/m}^3$ of total dust exposure were measured for chimney sweepers across their most common tasks. Exposure to BaP varied from below the detection limit to $9.1 \,\mu\text{g/m}^3$ (for an average of $53 \,\text{min}$) (Andersson 1987). Another exposure assessment from Sweden showed that chimney sweepers were exposed to a median inhalable dust concentration of $3.8 \,\text{mg/m}^3$ during an 8-hr working day while sweeping in private homes (Hogstedt et al. 2013).

Nanoparticles are particles with at least one dimension measuring <100 nm (European Committee for Standardization 2018). Exposure to nanoparticles can cause adverse health effects, as their small size allows translocation through the air-blood barrier and direct interaction with systemic circulation, meaning that nanoparticles can reach organs (Yacobi et al. 2011; Elsaesser and Howard 2012; Liu et al. 2021). Focus on nanosized particles has been rapidly increasing due to the elevated production and use of engineered nanomaterials, and several portable nanosized particle counter instruments are now available for assessing personal exposure. Chimney sweepers may also be exposed to high concentrations of nanoparticles, considering that soot from wood combustion contains large amounts of the nanometer size fraction (Trojanowski and Fthenakis 2019). However, there is no OEL for exposure to nanosized particles in the work environment, even though these particles have been found to drive inflammation and compared with larger particles of the same chemistry, produce more significant inflammatory responses due to their greater surface areas per unit mass (Brown et al. 2001; Oberdörster et al. 2005).

At an International Workshop on Nano Reference Values organized by the Dutch trade unions and the Social Economic Council in 2011, two occupational nano reference limit values were proposed, namely, 2.0×10^4 particles/cm³ for bio-persistent, granular nanomaterials (1–100 nm) with densities >6,000 kg/m³, and 4.0×10^4 particles/cm³ for bio-persistent granular nanomaterials and their fibers (1–100 nm) with densities <6,000 kg/m³ (8-hr TWA), respectively (Van Broekhuizen et al. 2012). Considering that the density of soot is less than 6,000 kg/m³, the reference value of 4.0×10^4 particles/cm³ (8-hr TWA) can be used when assessing exposure to nanosized particles in soot.

Additionally, there is another, possibly more relevant, metric for measuring the adverse health effects

of aerosol particles. This metric combines the lung deposition of particles and the potential for surface chemistry and is referred to as the lung-deposited surface area (LDSA). LDSA is defined as the particle surface area concentration per unit volume of air, weighted by the deposition probability in the lung. The deposition probability is customarily calculated according to the ICRF report 66 (ICRP 1994). LDSA is introduced as an answer to the toxicological idea that the particle surface area in the lung matters for measuring exposure (Fierz et al. 2011).

In recent years, occupational exposure among chimney sweepers has decreased with the implementation of designated contamination zones, increased use of personal respiratory equipment, and increased knowledge of potential health effects caused by exposure. However, limited studies on airborne exposure to PAHs in gaseous and particulate phases have been conducted among chimney sweepers, and to the best of the authors' knowledge, exposure to nanoparticles among chimney sweepers has not been systematically measured.

This study aimed to assess the time-weighted average (TWA) of airborne exposure to gaseous and particulate PAHs, along with the particle number concentrations (PNCs), particle sizes, and LDSAs, for chimney sweepers in this region of Norway. Additionally, it aimed to assess the average, with the standard deviation (SD) and the range of PNCs and LDSAs generated across different tasks. Exposure was first evaluated during a "typical work week" of chimney sweeping and fire safety inspections, then during a "massive soot" week, during which larger sweeping jobs took place.

Methods

Recruitment of study participants

The recruitment of chimney sweepers was facilitated by reaching out to their central workplace leader. Subsequently, a meeting involving the leader, safety representative, chimney sweepers, and researchers was conducted, during which the researchers informed them about the study. Additionally, written information ensuring clarity about the study's objectives, procedures, participant rights, and potential outcomes was sent out before the sampling began. Criteria for participation were (a) working as a chimney sweeper, and (b) being 18 years old or older. The chimney sweepers gave oral consent to wear the sampling equipment. This study involved monitoring employees in a workplace where health and safety are considered the leader's responsibility. For this reason, the study fell outside Norwegian health research legislation, making ethics committee approval for health research unnecessary. Furthermore, the samples were anonymized for publication to meet privacy and security laws and requirements.

Sampling strategy

Samples were collected over 2 weeks: 1 week (five working days) in November 2021 (8-12 November), followed by 1 week (four working days) in March 2022 (14-17 March). Due to the researchers' access to sampling pumps, it was decided to follow two teams of four chimney sweepers each. However, due to sick leaves and new trainees, the team compositions changed slightly during the sampling period. Of the 29 chimney sweepers working out of their central workplace, 17 participated in this study. On a typical day, two sweepers on a team swept the chimneys from the attic or outdoors from the roof, then collected the soot from the soot hatches inside individual family homes or the basements of larger residential blocks. The remaining two sweepers on the team conducted fire safety inspections. Each workday started at 7:00 AM, and planned sweeping activities were performed from approximately 8:00 to 11:30 AM. After lunch, the sweepers participated in different activities, such as unplanned/emergency sweeping, exercising, and planning sweeping activities for the week ahead. In the second week of sampling, sweeping jobs took place mainly indoors in commercial locations, including a large indoor waste sorting facility, restaurant ovens, and tiled stoves, all with potentially higher exposure concentrations. The sweepers called this week a "massive soot" week, as the tasks were more extensive than those conducted during a typical work week. "Massive soot" weeks are performed for approximately 3 months per year.

Using daily work logs, the chimney sweepers recorded information on their tasks, which was then uploaded into their central workplace digital logging system at the end of their shifts. The total number of sweeping jobs/fire safety inspections and the estimated times spent on the tasks across the 2 weeks of sampling were collected from this system. Additionally, to connect the patterns observed in the real-time sample information on PNC, size, and LDSA with the tasks performed, one to three researchers joined the sweepers in the field each day to make detailed notes on tasks and the time spent on each activity.

Measures to reduce exposure

Many measures to reduce chimney sweepers' exposure have been implemented at their central workplace, including the designation of contamination zones (dirty, transitional, clean) to control the spread of harmful contaminates in the central workplace and vehicles, and the use of personal protective equipment (PPE), including respirators with gas and particle filters, gloves, long-sleeved jackets, and long pants. When necessary, the chimney sweepers also use overalls and goggles. For respirators, most of the chimney sweepers in this study used powered air purifying CleanSpace2 half-facepiece reusable respirators (PAF-0034; CleanSpace Technology Pty Ltd, 16-18 Carlotta Street, Artemon NSW 2064, Australia) with combined filters (A1P3). Some used powered air purifying CleanSpace full face masks (PAF-1014) during tasks where exposure levels were expected to be high.

Assessing exposure

Between one and eight personal samples, mounted in the breathing zone on the outside of the respirators, were collected from each of the 17 chimney sweepers in this study. In total, 70 samples of particulate PAH were collected. Additionally, in series with 28 of the particulate PAH samples, adsorbent tubes were connected to collect information on the concentration of gaseous PAH. The concentrations of particulate PAHs were measured using closed-face filter cassettes containing 37 mm Teflon filters/PTEF membrane filters, with a pore size of 2 µm (SKC 225-1713). Gaseous PAHs were collected on XAD2 adsorbent tubes (SKC 226-30-04), which were coupled in series with the Teflon filters, following the protocol in NIOSH NMAM 5515. The samples were connected to a sampling pump (Casella Apex2), which was calibrated to 2.0 L/min. The flow through both sample trains (particulate PAH only, and the particulate and gaseous PAH samples collected in series) was verified both before and after sampling. The flows were stopped when the sweepers were done with their sweeping activities for the day so that they would not need to sit with the sampling equipment while in the office or while exercising. The flows therefore ran between 2.3 and 7.5 hr each day of sampling. The office/exercise areas at their central workplace were clean zones, and the sweepers were not considered to be exposed to PAH while working in the central workplace. After sampling, the XAD2 tubes were wrapped in aluminum foil, and together with the filters and field blanks, were sent to the NS-EN ISO 17025 accredited lab Nemko Norlab, where the PAHs collected onto the filters and XAD2 tubes were extracted

separately, and internal standards (isotopically labeled PAHs) were added before processing. All PAH samples (gaseous and particulate) were analyzed using gas chromatography-mass spectrometry (GC-MS). In addition to naphthalene and biphenyl, the lab reported the following 18 PAHs, in gaseous and particulate phase: phenanthrene, anthracene, fluoranthene, pyrene, benzo(a)fluorene, benzo(b)fluorene, chrysene, benzo(a)anthracene, benzo(bjk) fluoranthene, benzo(e)pyrene, BaP, dibenz (a,h)anthracene, indeno(1,2,3,-cd)pyrene, benzo(ghi)perylene, dibenzo(ah)pyrene, dibenzo(ae)pyrene, dibenzo(ai)pyrene, and dibenzo(al)pyrene.

Biphenyl and naphthalene were reported separately, while the remaining PAHs were reported both individually and combined. According to the lab, the detection limit for the sum of the remaining PAHs, biphenyl, and naphthalene was $0.001 \,\mu\text{g/m}^3$. For single PAHs below this limit, the quantification limit was halved before being interpreted for the sum.

Additionally, on each sampling day, one chimney sweeper wore a DiSCmini nanoparticle counter (Testo, Germany) for real-time personal monitoring of PNCs which was placed in the breathing zone, above the respirator, of each worker. The DiSCmini is based on unipolar diffusion particle charging and detection done in two stages by electrometers and determines PNCs and average particle sizes in the range of 10-300 nm. The LDSA considers the depositional efficiency of particles in different lung compartments (ICRP 1994) and is calculated directly by the instrument. The time resolution of the instrument is 1 sec, with a reading accuracy of $\pm 30\%$ (Fierz et al. 2011). The calculation and LDSA are, in principle, based on the product of fraction deposition and particle mass in each size range (Fierz et al. 2011, 2014; Vora et al. 2021). The instrument was cleaned and zero-checked by connecting it to a HEPA filter before each measurement.

Calculating exposures

To compare the concentrations of PAHs against the Norwegian OELs, along with the nano reference value for PNCs, the concentrations were normalized (TWA) to an 8-hr reference period.

In cases where the sample duration of PAH was less than 8 hr, zero exposure was assumed for the remaining time as the time was spent in the office or exercising (clean zones). In the clean zones of the station, PAH concentrations were assumed to be negligible. For the values measured using the DiSCmini (PNC, LDSA, and particle size), the average values measured in their central workplace (clean zone) in the morning before sweeping were used as the "background" concentrations for T_2 .

For the PNC, size, and LDSA concentrations reported per task, the average, SD, and range were calculated from the 1-sec averages in Excel, and the concentrations measured over task times were coupled with the time logs made by the researchers in the field.

Statistical analysis

The Shapiro–Wilk test was used to evaluate the normality of the concentration data, both log-transformed and non-transformed, and the data were not normally distributed. For this reason, the nonparametric Mann–Whitney U test was used to test for possible differences in gaseous- and particulate-phase PAHs and PNCs across the 2 weeks. Spearman's correlation (ρ) was used to assess possible correlations between the TWA of PAHs and PNCs. Statistical analysis was performed using IBM's SPSS 28.

Results

Two particulate PAH samples were excluded, one due to cassette leakage and the other due to a cassette falling off early during the day. One real-time sample of nanoparticles was excluded, as the logger accidentally stopped before the sweeper started working. As such, 68 particulate PAHs, 28 gaseous PAHs, and 8 realtime samples of nanoparticles were available for analysis.

According to the chimney sweepers, a typical workday consisted of each team sweeping 10–80 chimneys depending on chimney size and building type. During the first week, some sweepers only did fire safety inspections, while most alternated between fire safety inspections and sweeping. Based on observations made during the first week of sampling, sweeping the roof chimney of a single-family building took 2–3 min, while soot collection in private homes took approximately 10 min. For large apartment buildings, where several fireplaces were connected to the same chimney, sweeping one chimney took about 10– 15 min, followed by soot collection, where emptying each soot hatch took about 10 min.

Based on the log schemes collected from the chimney sweepers, the self-reported mean time spent on different tasks per day is reported in Table 1. As shown, November's average time spent on chimneys sweeping per day was 3.6 hr (SD = 3.4). During the "massive soot" week in March, the average time spent on chimney sweeping increased to 5.2 hr (SD = 4.8) per day. Eleven chimney sweepers also logged their activity between the two sampling weeks, from November to March, and the average number of chimney sweepings per day in this period was 13.9 (SD = 11.8), comparable with the first week of November.

Table 2 shows the TWAs of the gaseous- and particulate-phase PAHs, biphenyl, and naphthalene. As shown, slightly higher concentrations of PAHs were measured in the gaseous phase compared to the particulate phase. Significantly higher concentrations of PAHs, both in the gaseous phase and in the particulate phase, were measured during the "massive soot" week in March 2022, compared to the typical work week in November 2021 (Mann–Whitney U test, p < 0.001).

During the first week of sampling, gaseous BaP was quantified in 3 out of 10 samples, while particulate BaP was quantified in 11 out of 37 samples. During the second week, BaP was quantified in 15 out of 19 and 16 out of 32 samples in its gaseous and

 Table 2. TWA (8 hr) and the number of samples collected of the gaseous and particulate PAHs.

	No	vembei	r 2021	March 2022					
	Mean*	Min	Max	Ν	Mean*	Min	Max	Ν	
Total PAH _{particle}	0.65	0.49	2.50	36	1.09	0.87	1.39	32	
Biphenylparticle	0.01	0.00	0.01	36	0.00	0.00	0.36	32	
Naphthaleneparticle	0.13	0.00	0.23	36	0.02	0.00	0.41	32	
Total PAHgas	0.71	0.52	1.06	9	1.45	1.04	4.47	19	
Biphenylgas	0.00	0.00	0.01	9	0.01	0.01	0.02	19	
Naphthalene _{gas}	0.07	0.02	0.13	9	0.04	0.00	0.09	19	

*Concentrations in µg/m³.

 Table 1. Self-reported mean time (SD) spent on different tasks per day in the 2 weeks of sampling.

Period/activity	Number of chimney sweeps (SD)	Time (hr) spent on chimney sweeping (SD)	Number of fire-safety inspections** (SD)	Time (hr) spent on fire- safety inspections (SD)
November 8–11, 2021*	14.3	3.6	9.9	2.7
	(16.8)	(3.4)	(12.9)	(2.9)
March 14–17, 2022*	18.1	5.2	7.4	1.2
	(17.8)	(4.8)	(22.1)	(3.4)

*15 and 17 chimney sweepers reported their time spent on different activities in November and March, respectively.

**Inspection of fireplaces and soot hatches.

particulate phases, respectively. Normalized to an 8-hr shift, the TWA of BaP, summarizing the concentrations measured in the gaseous and particulate phases, ranged from <0.001 to $0.087 \,\mu\text{g/m}^3$. A positive Spearman correlation was observed between gaseous and particulate PAHs ($\rho = 0.61$, p < 0.001, n = 28), while no correlation was observed between PAHs and PNCs. However, this latter correlation was limited to eight pairings due to limited PNCs.

As shown in Table 3, the TWAs of the particles varied from 2.0×10^3 to 7.1×10^4 particles/cm³. As for the PAHs, the TWA for the PNC was significantly higher (p < 0.05) in the second week than the first, with medians of 4.2×10^3 and 2.2×10^4 particles/cm³, respectively. The lowest concentration of particles was measured on a day when a chimney sweeper was exposed to soot only for three short periods of 4, 8, and 12 min. On this day, up to 8.7×10^4 particles/cm³ were measured (see Table 4). However, the sweeper was not exposed to soot for the remainder of the day, resulting in a TWA at the same level as expected in the office area $(2.0 \times 10^3 \text{ particles/cm}^3)$. As shown in Table 3, the highest PNC TWA was measured during indoor sweeping in the waste sorting facility. This work was mainly divided between two and three chimney sweepers and was conducted over 2 days

Table 3. TWAs for PNC, LDSA, and particle size across different sampling days.

Week	Date	PNC (particles/cm ³)	Size (nm)	LDSA (µm²/cm³)
1	November 8, 2021 November 10, 2021	$\begin{array}{c} 4.4\times10^3\\ 2.0\times10^3\end{array}$	49 42	10.5 3.5
-	November 11, 2021 November 12, 2021	9.6×10^{3} 4.0×10^{3}	46 34	14.4 6.8
2	March 14, 2022 March 15, 2022 March 16, 2022	3.7×10^{3} 7.1×10^{4} 2.6×10^{4}	52 44	9.1 54.4
	March 17, 2022	3.0×10^{3} 8.1 × 10 ³	40 50	18.2

(March 15-16, 2022), where the same worker, a trainee, carried the DiSCmini nanoparticle counter, as he was considered the most exposed person. As shown in Table 4, during the 2 days of sweeping in the waste sorting facility, the PNC TWAs were 3.6×10^4 and 7.1×10^4 particles/cm³ for Day 1 and Day 2, respectively, with LDSAs of 49.4 and 54.4 μ m²/cm³, respectively. However, looking only at the periods spent sweeping inside the facility, which lasted 120-160 min, the average PNCs reached approximately 1.0×10^5 and 2.1×10^5 particles/cm³ for Day 1 and Day 2, respectively, with LDSA concentrations of between 131.4 and 141.9 µm²/cm³, and average particle sizes in the range 14.6-21.8 nm, respectively. On the second day, when the highest number of particles was measured, the worker crawled into a narrow tube to sweep and vacuum. While engaging in this activity, he wore a coverall taped around all openings to reduce skin exposure. Also, he was wearing protective respiratory equipment, gloves, and goggles.

In addition to carrying the nanoparticle counter for the 2 days spent in the waste sorting facility, the same worker carried it on a third day while sweeping inside a tiled stove (17 March 2022), and the average TWA of exposure for these 3 days was 3.8×10^4 particles/cm³.

In Table 4, the particle concentrations for different tasks are summarized. As shown in the table, chimney sweeping, or sweeping pizza ovens and tiled stoves, produced an average of $6.8 \times 10^3 - 6.3 \times 10^4$ particles/ cm³ for between 25 and 80 min. Dismantling a fire-place produced an average of 2.7×10^4 particles/cm³, lasting 50 min. During chimney sweeping and dismantling, the mean particle size varied from 27.8 to 56.9 nm, and the LDSA concentration varied from 17.0 to 80.7 μ m²/cm³. During all soot-generating activities, the sweepers wore long-sleeved work clothes, respiratory protective equipment, gloves, and sometimes overalls and goggles.

Table 4.	The arithmetic mean	(AM), SD and	range of PNCs, s	ize, and LDSA	measured during	different soot-a	enerating
i alore iii	The analiticate mean	(<i>n</i> in <i>n</i>) = 50 and	runge of fittes, s		measured during	anicient soot g	chickating

		Time	PNC (particles/cm ³)				Size	(nm)	LDSA (µm ² /cm ³)		
Date	Activity	(min)	AM	SD	Min – Max	AM	SD	Min – Max	AM	SD	Min-max
November 8, 2021	Chimney sweeping	25	1.3×10^4	1.3×10^4	$2.3\times10^39.4\times10^4$	42.2	5.4	21.0 - 69.7	26.6	20.2	7.3 – 149.9
November 10, 2021	Chimney sweeping	35	$1.4 imes 10^4$	$1.2 imes 10^4$	0.8×10^{3} - 8.7×10^{4}	27.8	9.1	10.0 - 56.1	19.6	15.9	1.7 — 108.6
November 11, 2021	Chimney sweeping	40	4.3×10^4	$1.0 imes 10^5$	0.5×10^{3} -7.8 $\times 10^{5}$	36.8	32.2	10-300*	42.2	79.2	2.3 - 814.3
	Dismantling of fireplace	50	$2.7 imes 10^4$	$1.1 imes 10^5$	$1.0\times10^32.8\times10^6$	38.6	37.0	10-300*	50.8	178.5	4.4 – 4499.1
November 12, 2021	Emptying the vacuum cleaner	10	$6.3 imes 10^4$	$2.7 imes 10^5$	$2.0\times10^32.9\times10^6$	29.6	21.2	10-300*	109.7	459.2	14.6 - 4547.0
March 14, 2022	Sweeping and emptying the soot hatch of a pizza oven	45	$6.8 imes 10^3$	$1.6 imes 10^4$	0.2×10^{3} - 6.3×10^{5}	56.9	34.4	10–300*	17.0	25.3	3.0 - 922.3
March 15, 2022	Sweeping in a waste sorting facility	160	$2.1 imes 10^5$	8.2×10^4	$3.1 imes 10^{3} extrm{-}7.6 imes 10^{5}$	14.6	7.8	10.0 - 67.6	141.9	53.1	7.0 — 605.1
March 16, 2022	Sweeping in a waste sorting facility	120	$1.0 imes 10^5$	$1.9 imes 10^5$	$2.6\times10^32.9\times10^6$	21.8	16.6	10-300*	131.4	347.8	7.6 – 4548.2
March 17, 2022	Sweeping in a tiled stove	80	$3.6 imes 10^4$	$7.9 imes10^4$	$0.18\times10^3{-}1.7\times10^6$	47.5	19.1	10-300*	80.7	162.0	1.1 — 1659.7
	and milling										

*The DiSC mini reads average particle sizes in the range 10-300 nm.

Discussion

Overall, the PAH concentrations and PNCs were significantly higher during "massive soot" week compared to a typical week of chimney sweeping and fire safety inspections. However, these findings suggest that the chimney sweepers included in this study were not exposed to high concentrations of PAHs while sweeping, even when the use of respiratory protection was not factored into the exposure analysis. Additionally, based on conversations and observations, the necessity of using PPE, such as respiratory protection, long pants and jackets, gloves, and when necessary, overalls, and goggles was understood. However, the PNCs were high during certain activities, including emptying the vacuum cleaner (limited to 10 min) and during the 2 days of sweeping in the waste sorting facility, reaching PNC TWA levels of 3.6×10^4 and 7.1×10^4 particles/cm³, respectively, across the 2 days. These concentrations are near and above, respectively, the nano reference limit value of 4.0×10^4 particles/cm³ proposed by the International Workshop on Nano Reference Values (Van Broekhuizen et al. 2012). Many previous studies have reported health outcomes associated with chimney sweeping and the concentrations of various PAH metabolites after sweeping. However, studies reporting the actual exposure concentrations while sweeping have been limited. To the best of the authors' knowledge, this is one of the first studies reporting the airborne concentrations of gaseous and particulate PAHs for chimney sweeping and the first study reporting the PNCs for this occupation.

Exposure to the gaseous- and particulate-phase PAHs

In this study, the TWAs of gaseous and particulate PAHs measured ranged from 0.52 to $4.47 \,\mu\text{g/m}^3$ and 0.49 to $2.50 \,\mu\text{g/m}^3$, respectively. When summarizing the gaseous and particulate PAHs (excluding naphthalene and biphenyl) measured in series (n = 28), the PAH TWAs ranged from 0.81 to $5.63 \,\mu\text{g/m}^3$, well below the Norwegian OEL of 40 µg/m³. When summarizing the gaseous- and particulate-phase BaPs (n=28), the TWAs ranged from below the detection limit (<0.001) to 0.087 µg/m³. In Norway, no OEL for BaP exists, however, the TWA of BaP measured in this study was below the Swedish OEL (8 hr) of $2 \mu g/$ m³ (Swedish Work Environment Authority 2018). Compared to the Dutch OEL, which is based on an estimated excess cancer risk of 4 per 10⁵ across 40 years of occupational exposure, 14 of the 28

summarized gaseous and particulate BaP samples, and one of the particulate BaP samples, exceeded the OEL of 5.7 ng/m^3 (DECOS 2006).

The concentrations measured in this study are challenging to compare with those reported in previous studies on PAH exposure among chimney sweepers, as TWAs were not usually calculated in previous studies. Although the PAH concentrations in this study were below the Norwegian OEL, results can be compared with the PAH concentrations measured among other occupations also exposed to soot. The chimney sweepers included in this study were exposed to higher concentrations of PAHs compared to chefs in three Norwegian ala-carte restaurants, where exposure to naphthalene, the only PAH detected, was measured in the range of $0.05-0.27 \,\mu\text{g/m}^3$ (Sjaastad and Svendsen 2009). Additionally, the chimney sweepers in this study were also exposed to higher PAH concentrations compared to Italian asphalt and construction workers, where the median air concentrations of PAHs ranged from <0.00005 to $0.43 \,\mu\text{g/m}^3$ (Cirla et al. 2007).

Control measures among the chimney sweepers

In a Swedish study, 483 chimney sweepers were asked about their occupational history and eye and airway symptoms using a questionnaire. The results showed that the mean percentage of chimney sweepers using gloves, respiratory protection, and vacuum cleaners while working with black soot significantly increased in the period 1975 to 2010 (Alhamdow et al. 2017). In this study, chimney sweepers always used protective respiratory protective equipment with filters for gas and particles and sometimes also a powered-air purifying respirator, long-sleeved suits, and gloves, and when necessary, coveralls and goggles. During the weeks of observation, no discussions regarding the necessity of PPE occurred, and the workers seemed to be aware of the potential health effects of exposure to soot. However, emptying the vacuum cleaner was not considered an activity where exposure would occur, and no PPE was used during this task. As shown in Table 2, this 10-min task generated, on average, 6.3×10^4 particles/cm³, with an average particle size of 29.6 nm and an LDSA concentration of 109.7 $\mu m^2/cm^3$.

Presumably, with the implementation of measures such as designated contamination zones, increased use of PPE, and increased knowledge of potential health effects caused by exposure, the concentrations that the chimney sweepers are exposed to have decreased, and older published results on the prevalence of health impacts among chimney sweepers may no longer be equally relevant. Nevertheless, although limited studies on air exposure to PAHs among chimney sweepers exist, the results of a recent study, where PAH metabolites were measured among chimney sweepers, creosote-exposed workers, and controls, up to three times higher median concentrations of PAH metabolites were found in the urine of chimney sweepers compared to the controls (Alhamdow et al. 2020). Additionally, chimney sweepers had up to seven times higher concentrations of PAH metabolites in their urine than the controls (p < 0.001) (Alhamdow et al. 2017). Chimney sweepers are also exposed to phenanthrene and fluorine with the latter being associated with DNA hypomethylation of F2RL3 and AHRR, prospective markers of lung cancer (Alhamdow et al. 2020). This finding suggests that even though various measures to reduce the exposure concentrations have been implemented, chimney sweepers may still be exposed to unacceptably high concentrations of PAHs at work.

Exposure to nanosized particles

As shown in Table 3, the PNCs measured in the waste sorting facility were near and above the proposed nano reference value of 4.0×10^4 particles/cm³ (8-hr TWA) (Van Broekhuizen et al. 2012). Although only one worker was carrying the DiSCmini daily, the other sweepers sweeping in the waste sorting facility were assumingly also exposed to high PNCs, as they helped each other with the different work tasks.

Instead of using PNC and particle size, LDSA has been used as a measure of exposure in some previous studies. In a study from Finland, the average LDSA concentrations in urban air varied from 12 (park area) to 94 μ m²/cm³ (major road) (Kuuluvainen et al. 2016). In another study characterizing the exposure to airborne nanoparticles across taconite mine operations, the concentrations of ultrafine particles were measured during crushing, dry milling, wet milling, and pelletizing. The results showed that the highest LDSA concentrations were generated during pelletizing (199±48 μ m²/cm³), followed by crushing (141±52 μ m²/cm³), dry milling (91±9 μ m²/cm³), and wet milling (85±7 μ m²/cm³) (Afshar-Mohajer et al. 2020).

Further knowledge about PNC TWAs in occupational settings can be gleaned from studies using other nanoparticle counters. In one study, a TSI P-Trak was utilized to determine the exposure of asphalt workers during paving, and peak concentrations frequently reached 1×10^5 to 2.1×10^5 particles/cm³ (Elihn et al. 2008). Also utilizing a TSI P-Trak, another study investigating paving and related road construction activities determined TWAs ranging from 2.3×10^4 to 7.0×10^4 particles/cm³ during paving, from 1.9×10^4 to 1.1×10^5 particles/cm³ during milling, and 3.6×10^4 to 4.5×10^4 particles/cm³ during pothole repair (Freund et al. 2012). In another study, the TWA among all studied furnace tappers was determined to be 1.74×10^4 particles/cm³ and varied between 1.47×10^4 and 2.06×10^4 particles/cm³ for different groups of tappers (Jørgensen et al. 2020).

Many studies have been performed using activitybased measurements. One of the highest exposure levels was found to occur during PVC welding, with PNCs of 3.3×10^6 particles/cm³ and a maximum peak of 8.1×10^7 particles/cm³ (Jørgensen et al. 2016). Based on these results, chimney sweepers are exposed to airborne nanoparticles at comparable levels to other highly polluted industries during the days of greatest exposure, while the average exposure concentrations on days with limited chimney sweeping are comparable with background levels ($2.67 \pm 1.79 \times 10^3$ particles/cm³) (Morawska et al. 2008).

Limitations

This study was relatively short, including a limited number of chimney sweepers and samples, and the results may therefore not be generalized across the entire chimney sweeper population. The focus of this study was on air exposure to PAH and nanoparticles among chimney sweepers. However, chimney sweepers might encounter exposure not exclusively via inhalation, as dermal contact could also represent a significant exposure pathway (Liang et al. 2013). In this study, the visible dermal exposure to soot varied across days, and on some days, exposed areas, such as the wrists, neck, face, and hair, were covered in soot. This may indicate that despite using PPE such as suits and long pants, gloves, and respiratory protection, chimney sweepers may face significant dermal exposures. Hence, measuring air exposure may not be sufficient to characterize their total exposure to PNCs and PAHs.

Portable nanoparticle monitors such as the DiSCmini may not be 100% accurate. However, utilization of the DISCmini presents the only reasonably feasible method for establishing knowledge about exposure to nanoparticles for chimney sweepers and similar occupations. Recent studies report overall good agreement between portable and stationary nanoparticle counters (Viana et al. 2015; Bémer and Bau 2019). Additionally, real-time samplers offer several advantages such as: easy identification of sources contributing to the exposure, visualization of variations in exposure to share with and motivate workers, and the determination of appropriate control measures that when implemented can reduce exposure concentrations (Moazami et al. 2023). For this study, the DISCmini portable instrument provided new insights into the exposure to nanosized particles and tasks contributing to the highest PNCs and LDSA concentrations measured.

Conclusion

For the chimney sweepers included in this study, the median TWAs (8 hr) for air exposure to gaseous and particulate PAHs were 0.52-4.47 and $0.49-2.50 \,\mu\text{g/m}^3$, respectively, well below the Norwegian OEL of 40 µg/ m³ for 21 PAHs. However, significantly higher concentrations of PAHs were measured during "massive soot" week, where more extensive sweeping activities were performed, compared to a typical week of chimney sweeping and fire safety inspections. Also, slightly higher concentrations of gaseous PAHs were measured compared to particulate PAHs. The chimney sweepers seemed aware of the potential health risks associated with exposure, and suitable PPE was used. However, the PNCs were high during certain activities, including emptying of the vacuum cleaner (limited to 10 min) and during the 2 days of sweeping in the waste sorting facility, reaching PNC TWA levels of 3.6×10^4 and 7.1×10^4 particles/cm³, across the 2 days, respectively, which are near and above, respectively, the proposed nano reference limit value of 4.0×10^4 particles/cm³. During sweeping in a waste sorting facility, an average PNC of up to 2.1×10^5 over 160 min was measured. The PNC and LDSA values reported per activity show that if the activities change or increase, the TWAs (8 hr) of nanosized particles can reach above 4.0×10^4 particles/cm³.

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Disclosure statement

The authors have no competing interest to declare.

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Data availability statement

The data supporting the findings of this study are available from the corresponding author, T.N.A., upon reasonable request.

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