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Removal of polycyclic aromatic hydrocarbons and genotoxic compounds in urban air using air filter materials for mechanical ventilation in buildings

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Humans spend most of their lives in indoor environments; hence, indoor exposure to air pollution may constitute a large part of the total exposure to air pollution. Polycyclic aromatic hydrocarbons are well known for their mutagenicity and carcinogenicity and are ubiquitous in urban environments as a result of combustion from e.g. vehicular traffic. Polycyclic aromatic hydrocarbons associated to air particulate matter in indoor environments originates from several sources including: cooking and heating, outdoor sources, smoking, candle and incense burning. Infiltration has been suspected to be one major source of indoor polycyclic aromatic hydrocarbons. In this study, four different air filter materials intended for mechanical ventilation were tested for their capability to remove particle bound polycyclic aromatic hydrocarbons and other genotoxic compounds from a real urban aerosol. Particles were sampled at two highly trafficked locations in Stockholm using a sampling system capable of sample particles in parallel, thus allowing sampling of filtered and un-filtered air simultaneously. The sampled particles were extracted and analysed for polycyclic aromatic hydrocarbons and the genotoxicity of the organic extract was determined using Ames mutagenicity tests. Each air filters capability of removing polycyclic aromatic hydrocarbons and reducing genotoxic effects was determined by comparing the filtered and un-filtered air samples. The results showed that all air filter materials had the capability of removing polycyclic aromatic hydrocarbons and reduce genotoxic effects downstream the air filter, and that the magnitude of the reduction was correlated with the standardized particulate collection efficiencies of a 0.4 μm particles for the tested air filter materials. However, the filter with the lowest performance did not significantly reduce direct acting mutagens.

Introduction

Airborne particulate matter (PM), comprising the two common particle metrics of PM_{10} (aerodynamic diameter: <10 μm) and $PM_{2.5}$ (aerodynamic diameter: <2.5 μm), has been associated with several adverse health effects, including cardiovascular diseases and respiratory diseases (Bernstein et al. 2004; Brook et al. 2010; Kunzli et al. 2000). It has been estimated that exposure to $PM_{2.5}$ alone was responsible for 350,000 premature deaths in Europe in the year 2000 (Watkiss et al. 2005). Air pollution and PM have recently been classified as human carcinogens (Group 1) by the International Agency for Research on Cancer (IARC; Loomis et al. 2013), and particulate emissions from vehicles, i.e., tail pipe emissions and tire tread wear particles, are regarded as the major anthropogenic sources of PM in urban environments (Amato et al. 2013; Querol et al. 2004).

The cause of the adverse health effects from PM inhalation is still not fully understood, and it is still not clear whether it is the physical, e.g., particle size, or chemical properties that is of the highest importance (Harrison and Yin 2000). Nevertheless, one group of compounds that has been suggested to play an important role on the adverse health effects of PM inhalation is polycyclic aromatic hydrocarbons (PAHs; de Kok et al. 2006). PAHs are a large group of compounds consisting of two or more fused aromatic rings that are formed from incomplete combustion of organic material. PAHs are ubiquitous and persistent environmental contaminants that are both mutagenic and carcinogenic (Boström et al. 2002; Lee et al. 1981). Vehicular exhaust emissions have been regarded

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as a major source of airborne PAHs in urban environments (Boström et al. 2002; Jang et al. 2013). PAHs in ambient air with molecular weight exceeding 228 Da are almost entirely associated with particles, whereas PAHs with lower molecular weights, such as phenanthrene (178 Da) and pyrene (202 Da), exist partitioned between its condensed state associated with particles and the gaseous phase (Jongeneelen 2001; Rosell et al. 1991). Although most of the airborne PAH mass is partitioned to the gaseous phase (Landlová et al. 2014), the relative carcinogenic potency of PAHs differs widely between different derivatives (Boström et al. 2002), where the more potent PAHs are those with a higher molecular weight and ultimately those associated with PM. Recent studies have shown that the majority of the PAHs bound to air particles are associated with the PM_1 fraction (Landlová et al. 2014; Layshock et al. 2010).

The relative contribution of PAH exposure from indoor and outdoor sources is still poorly understood (Choi and Spengler 2014). It has been estimated that humans spend more than 80% of their time in indoor microenvironments (Maertens et al. 2004; Wallace and Williams 2005), implying that indoor PAH levels represent a large portion of the total PAH that humans are exposed to from air pollution. Indoor sources of PAHs include cooking and heating, outdoor sources, cigarette smoking, and candle and incense burning (Maertens et al. 2004). PAHs originating from outdoor sources through infiltration and cigarette smoking have been shown to be the major sources of PAHs in the indoor environments in Krakow, Poland (Choi et al. 2008). Li and coworkers (2005) measured indoor and outdoor concentrations of PAHs in the Chicago area and concluded that outdoor levels of heavy PAHs ($\geq$228 Da) correlated with indoor levels of heavy PAHs. In a review by Maertens and coauthors (2004), the importance of location was pointed out, *i.e.,* urban versus rural, over indoor smoking habits for PAH mass fractions in settled house dust collected at homes in the United States. Not only can the levels of contaminants in settled house dust be used as an indicator to indoor pollution levels, it is also an important route of human exposure, where toddlers are a particularly vulnerable group (Butte and Heinzow 2002; Whitehead et al. 2011).

Similar to the correlation of indoor and outdoor PAH concentrations, the indoor particle levels and characteristics are often governed by the ambient outdoor levels and characteristics (Jamriska et al. 2000; Quang et al. 2013). One method to control and reduce the particles entering the interior from the outdoor air is by means of mechanical ventilation with air filtration. Mechanical ventilation with filtration has been shown to significantly reduce indoor particle levels of $PM_{2.5}$ (Park et al. 2014), submicron particles (Jamriska et al. 2000), and both total suspended particulates (TSPs) and PM_{10} (Partti-Pellinen et al. 2000), suggesting that there would be some possible health benefits associated with air filtration as a result of lowered exposure. However, the scientific literature regarding possible health benefits from air filtration is rather scarce and was recently been reviewed by Fisk (2013).

Nevertheless, the possible health benefits of indoor air filtration by the means of recirculation in a wood smoke affected community was evaluated by Allen and co-workers (2011), where a reduction of the indoor $PM_{2.5}$ concentrations by 60% was achieved using a portable air purifier equipped with a HEPA filter. Based on the observed reduction of inflammatory biomarker concentration and improved endothelial function of the test subjects, the authors concluded that air filtration could reduce cardiovascular morbidity associated with air particle inhalation. In a recent study by Happo and co-workers (2014), several toxicological endpoints and chemical composition (including PAHs) of PM in an apartment with filtered air supply (using an F7 class filter) and outdoor air were compared. The authors concluded that the outdoor sources had a limited effect on the indoor air quality, suggesting that the air filtration had its intended impact on the indoor environment.

The ventilation requirements are defined by national authorities and differ between countries. In Sweden, the Swedish National Board of Housing, Building and Planning (Boverket), the Swedish Work Environment Authority (Arbetsmiljöverket), and the Public Health Agency of Sweden (Folkhälsomyndigheten) stipulate specific requirements regarding acceptable ventilation for different indoor environments (Arbetsmiljöverket 2009; Boverket 2011; Folkhälsomyndigheten 2014). In general, the minimum air exchange rate is determined by laws and guidelines, while limiting values exist for several compounds and particles for outdoor air in the European Union, there is no limit set for indoor air quality or to the supplied air in terms of particles associated or gaseous contaminants such as PAHs.

While air filter materials ability to remove particles is well known, to the best knowledge of the authors, there have not yet been any studies evaluating different air filter materials ability to remove PAH and reduce genotoxic effects. Hence, this study has aimed to obtain a quantitative measurement of different air filter materials with different performances and abilities to remove several commonly measured PAHs and other genotoxic compounds associated with particles from an air stream in a real urban setting.

Materials and methods

Solvents and chemicals

All solvents used for extraction and as the mobile phase during chemical analysis were of high-pressure liquid chromatography (HPLC) grade purchased from Rathburn Chemicals Ltd. (Walkerburn, Scotland). The ethanol 95% used for cleaning was from Kemetyl AB (Haninge, Sweden), the anhydrous dodecane (>99%) was from Sigma–Aldrich (St. Louis, MO, USA), and the dimethyl sulfoxide (DMSO) (99.9%) was purchased from Fisher Scientific. An extensive list with purities and manufacturer of the PAH standards used in this present study is available elsewhere (Sadiktsis et al. 2014).

All the chemicals used for the Ames mutagenicity test (between 98% and 99% purity) were purchased from Fisher Scientific. Quinacine and B[*a*]P were used as positive controls. The lyophilized liver S9 from Aroclor 1254-induced male Sprague Dawley rat was purchased from Moltox, NC, USA.

Preparation of sampling filters

T60A20 Pallflex filters (Pall Corp., NY, USA) were used for the collection of TSPs. Circular pieces of the filter sheets with a diameter 165 mm were cut out using a scalpel, hereafter referred to as samplers. The samplers were cleaned with ethanol, followed by acetone and dichloromethane. The samplers were dried at 150°C in an oven for 1 h followed by storage in a desiccator prior use.

Sampling locations and air sampling rig setup

The sampling of TSPs took place on the busy street Hornsgatan in southern urban Stockholm (59.31718, 18.04848) and alongside a freeway situated at suburban Midsommarkransens gymnasium in Stockholm (59.30429, 18.01787).

These sites were chosen because of the availability to conduct measurements and the anticipated mixture of air pollutions from traffic and other city activities. The measurement campaign was started at Hornsgatan and was later continued at the sampling site situated at Midsommarkransens gymnasium.

The air sampling system setup, depicted in Figure 1, consisted of two parallel test rigs that allowed simultaneous tests with and without the test object. Thus, each sampling of particles was performed on filtered and unfiltered air stream simultaneously.

The sampling system was equipped with a high-pressure fan, two test object holders, piping, holders for the samplers, holders for the polyurethane foam (PUF) plugs, two airflow/volume measurement devices, and two computers. The reason for using only one fan for both unfiltered air and filtered air was the limitations of the electrical power supply system (safety fuse) that only allowed one fan to operate. The airflow and the sampled air volume was measured and logged at each leg using a VPS-R150-P400 (VP-Instruments, Delft, The Netherlands) connected to a laptop computer. Pressure drops over the test objects were measured intermittently to indicate the actual filter loading by a SWEMA Air 300 (Swema AB, Farsta, Sweden). The test objects were clamped and sealed to the circular material holder. The open surface area (filtering area) of the test object was adjusted by covering an appropriate area with tape to simulate the typical air velocity through an air filter in real life (0.1–0.3 m s^{-1}).

The holders for the samplers and the PUF plugs for sampling of semi-volatile PAHs were made of stainless steel. However, the large volume of sampled air required for the Ames mutagenicity test led to exceeding the sampling capacity of the PUF plugs; hence, no PUF samples were further analyzed. The samplers were mounted in the sampler holders and connected to the PUF holders in a clean laboratory area prior to each test. The equipment was cleaned by wiping the disassembled sampler holder and the PUF holder with ethanol between each sampling occasion. The samplers collect all airborne particle size fractions (TSPs) with an efficiency of >98%. Sampling air velocities were within the range of 0.44–0.97 m s^{-1}, which corresponds to the 98%–99% collection efficiency for TSP of the samplers.

Sampled PM mass was determined on the samplers used at the sampling campaign at Midsommarkransens gymnasium by weighing desiccated samplers before and after sampling.

Air filter materials

The air filter materials (hereafter referred to as test objects) that were used in this investigation are commercially available. Four different types of air filter materials provided by Camfil AB (CM265, City-Flo F7, CM285, and CM295) were chosen to reflect the type of air filters used in general ventilation systems for offices, hospitals, public buildings, and dwellings. All types consisted of fibrous materials (glass) for particle filtration used in bag air filters. Typically materials used in this study are a higher density of fine fibers that gives higher collection efficiency. One test object was of a combination type (City-Flo F7) comprised of an additional active carbon adsorption layer suited for the removal of gaseous contaminants. The particle collection efficiency of the different test objects was in the range of 20%–80% at a 0.4-μm particle size, which correlates approximately with the collection efficiency of PM_1 according to the International Organization for Standardization's (ISO) Standard ISO/DIS 16890-1 (ISO 2015). These initial collection efficiencies correspond to the typical M6 (CM265), F7 (CM285 and City-Flo F7), and F9 (CM295) air filter classes according to Standard EN779:2012 (European Committee for Standardization [CEN] 2012). The gas phase adsorption efficiency for the combination material was 85% for toluene, which was determined in compliance with Standard ISO 10121-2:2013 (ISO 2013). The other filter materials were assumed to have a negligible gas phase adsorption. However, CM295 showed no adsorption when tested in accordance to Standard ASTM-D5742 (ASTM 2010).

The test objects were initially tested for particle collection efficiency and pressure drop at the rated velocity using the Standard EN779:2012 (CEN 2012) test methodology for fractional collection efficiency. For the in situ test, each test object was measured in three replicates; City-Flo F7 was the exception because of technical issues with the sampling equipment. The first sampling was carried out at Hornsgatan in March 2013. The following sampling sequence at Midsommarkransens gymnasium started in the middle of September 2013 and was finalized in the end of October 2013. The sampling in the real urban environment was conducted for each test object between 2 and 7 days, corresponding to a sampled air volume of roughly 900–5000 m^3. The required sampling time was estimated from PAH concentrations obtained from previous measurements and the expected particle penetration of the test objects.

PM sample extraction

The samplers were stored at –20°C prior to PAH analysis and the Ames mutagenicity test. The collected particles were extracted using pressurized fluid extraction using an ASE 200 accelerated solvent extractor system (Dionex Corporation, Sunnyvale, CA, USA) at a temperature of 200°C and pressure of 3000 psi (20.7 MPa) with a solvent mixture of toluene and methanol 9:1 (V/V); detailed instrumental settings are avail-

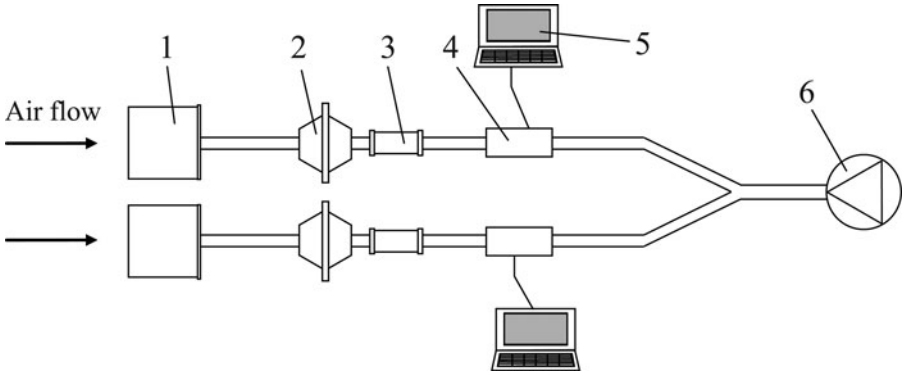

Fig. 1. Schematics of the air sampling system depicting: (1) test object holder, (2) sampler holder, (3) PUF holder, (4) airflow/volume measurement device, (5) computer, and (6) high-pressure fan.

able elsewhere (Masala et al. 2011). Cleaned samplers were used as blanks for the PAH analysis and Ames mutagenicity test.

The particulate extracts were collected in 60-mL preweighted vials. An aliquot was weighted (corresponding to roughly 1%–2% of the total weight of the extract) for PAH-analysis, while the remaining extract was used for Ames mutagenicity tests.

Preparation of PM extracts for Ames mutagenicity test

A volume of 500-μl DMSO was added to the raw extract as a keeper solvent. The extract was evaporated to roughly 5 mL using a TurboVap® LV evaporator (Zymark Corp., Hopkinton, MA, USA) at 70°C under a gentle stream of nitrogen.

The concentrated extract in the vial was transferred to a 7-mL flat bottomed test tube with screw cap. The walls of the vial were rinsed with hexane, which was added to the test tube. The extraction solvent was evaporated under a gentle stream of nitrogen gas and heating (54°C) using a heating block. The residual DMSO was filtered through a syringe filter (Titan3 30-mm Nylon 0.2 μm, SUN SRi, Rockwood, TN, USA) to remove the precipitates insoluble in DMSO. Another 500 μL of DMSO was used to rinse the small vial, which was also filtered in the same manner. The syringe filter was then rinsed with 500 μL of DMSO. The void volume of the syringe filters is <137 μl according to the product specification.

PAH analysis

An internal standard mixture containing perdeuterated PAHs (phenanthrene–d_{10}, pyrene–d_{10}, benz[*a*]anthracene–d_{12}, benzo[*a*]pyrene–d_{12}, and benzo[*ghi*]perylene–d_{12}) dissolved in toluene was added to the aliquot for PAH analysis and vigorously shaken using a vortex mixer. The extract was loaded on top of pre-conditioned (3 mL of hexane) silica SPE cartridge (Isolute IST, Biotage AB, Uppsala, Sweden) to remove polar constituents of the extract. The PAHs were eluted using 2 mL of hexane and the eluate was concentrated to less than 0.3 mL prior instrumental analysis.

A 50-μL aliquot of the DMSO extract for the Ames mutagenicity test was analyzed in a similar fashion as the raw extract. The determined native PAH concentrations were used to calculate the corresponding air volume per μl DMSO.

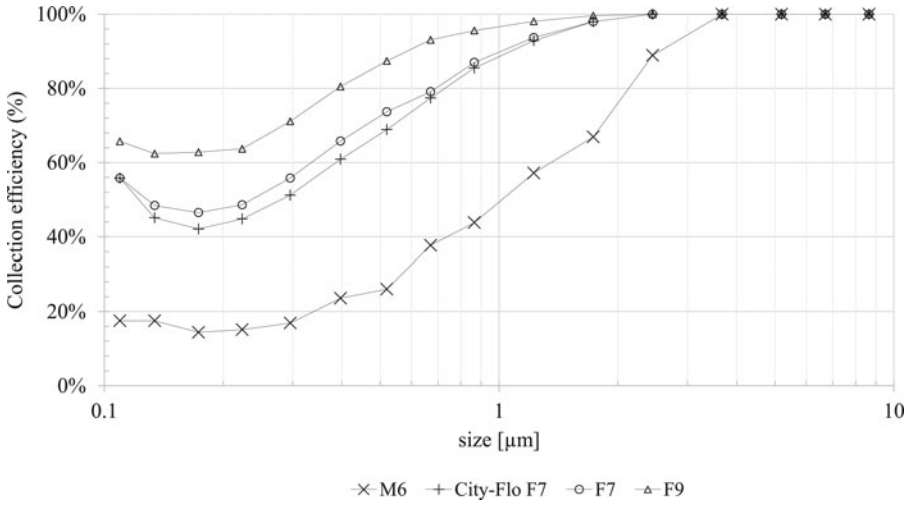

Fig. 2. Collection efficiencies of particles in the size range 0.1–10 μm for the four different test objects.

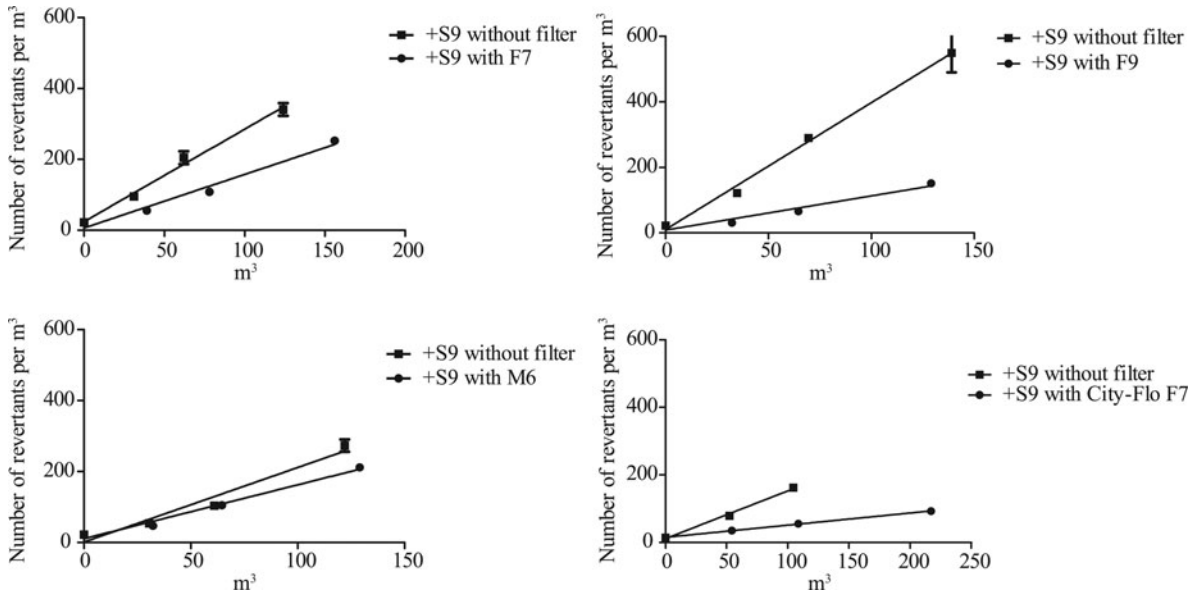

Fig. 3. Capability to reduce mutagenicity of air particles extracts by way of different air filter materials (F7, F9, M6, and City-Flo F7, respectively) exemplified by four different samples in the presence of a metabolizing system (+S9). The mutagenicity is given as the slopes (number of revertants per m^3) taken from the linear regression analyses. With all filter types, a significant reduction in mutagenicity was demonstrated (F7 and F9, $p < 0.0001$; M6, $p = 0.004$; City-Flo, $p = 0.002$).

The instrumental analysis was performed using a coupled HPLC with a gas chromatography mass spectrometer (HPLC-GC-MS) system. The HPLC part consisted of a Varian 9012 inert solvent delivery system (Varian Inc., Palo Alto, CA, USA) with a CMA/200 microsampler (CMA Microdialysis AB, Sweden). A PAH fraction with ≥3 rings were isolated using a back-flush HPLC (Östman and Colmsjö 1987) and introduced to the programmable temperature vaporizer injector inlet of the GC-MS system (Agilent 6890N gas chromatograph; Agilent Technologies, Palo Alto, CA, USA) coupled to an Agilent 5973N MSD system (Agilent Technologies). The GC separation was carried out on a DB-17MS capillary column (60 m × 0.25 mm inner diameter (i.d.) with a 0.15-μm film thickness with an integrated 5-m guard column; J & W Scientific, Folsom, CA, USA). The MS was operated in electron ionization mode (70 eV) with selected ion monitoring mode. Details on valve configurations and GC-MS parameters are available elsewhere (Christensen et al. 2005; Sadiktsis et al. 2014).

Ames mutagenicity tests

Mutagenicity tests of the DMSO soluble fractions of the particulate extracts were carried out using Salmonella typhimurium strain TA98 according to Maron and Ames (1983) with the modification given earlier (Westerholm et al. 2001). The volumes of the DMSO soluble fractions were adjusted to given concentrations (m^3 air μl^{-1} DMSO) comparable for all test objects. The soluble fractions were analyzed on plates with and without the metabolizing system S9. Each fraction was applied to plates at 0 (100 μl DMSO only), 25, 50, and 100 μl per plate in triplicates ($n = 12$) with and without 50 μl of S9. Linear regression analysis was then applied to the data using a GraphPad Prism 5.00 (GraphPad Software, Inc., La Jolla, CA, USA). Since sampling with the test air filter material was carried out simultaneously with the direct sampling without air filter material, it was possible to calculate the mutagenicity in terms of number of revertants per m^3 and to evaluate the capacity of the air filter to reduce the genotoxicity of the soluble particulate fractions. One-way analysis of variance (ANOVA) was conducted to compare the reduction efficiencies of mutagenicity of the different tested filter materials.

Results

Particle collection efficiency

The test objects were initially tested for particulate collection efficiency (Figure 2) in the size range of 0.1–10 μm in ac-

Table 1. Particle collection efficiencies of 0.4-μm particles, TSPs, and removal efficiencies of PM associated PAHs of the test objects (mean ± one standard deviation).

	Particle collection efficiency 0.4 μm, $n = 3$ (%)	Particle collection efficiency TSP, $n = 3$ (%)	Reduction efficiency of ∑PAH18, $n = 3$ (%)
F9	80.5 ± 1.3	90.4 ± 2.1	81.0 ± 2.7
F7	65.8 ± 1.8	87.0 ± 3.1[a]	65.5 ± 3.8[b]
City-Flo F7	60.9 ± 2.0	86.0 ± 0.4[a]	63.3 ± 4.4[a]
M6	23.6 ± 0.6	76.8 ± 9.8	37.5 ± 9.6

[a] $n = 2$.
[b] One measurement at Hornsgatan sampling site is included.

Table 2. Average, median, highest, and lowest air concentration of the individual PAHs measured at Midsommarkransens gymnasium ($n = 10$) and Hornsgatan ($n = 1$).

	Average concentration (pg/m^3)	Median concentration (pg/m^3)	Minimum concentration (pg/m^3)	Maximum concentration (pg/m^3)
Midsommarkransens gymnasium ($n = 10$)				
Phenanthrene	185	167	112	308
Anthracene	53.2	53.5	18.5	120
Fluoranthene	250	191	112	475
Pyrene	243	200	120	398
Benzo[c]phenanthrene	53.6	23.2	10.7	271
Benz[a]anthracene	131	105	50.5	251
Chrysene	183	161	81.2	340
Benzo[*b*]fluoranthene	242	229	54.8	451
Benzo[*k*]fluoranthene	114	103	23.6	221
Benzo[*e*]pyrene	188	175	55.8	311
Benzo[*a*]pyrene	179	170	49.9	297
Perylene	28.1	25.2	9.23	47.8
Indeno[1,2,3-*cd*]fluoranthene	18.9	17.7	2.86	43.2
Indeno[1,2,3-*cd*]pyrene	146	136	32.2	288
Dibenz[*a*,*h*]anthracene	29.6	26.5	4.72	69.7
Picene	27.3	25.0	4.19	63.2
Benzo[*ghi*]perylene	222	205	79.6	355
Coronene	120	121	58.8	186
$\sum$PAH18	2410	2320	1030	4140
Hornsgatan ($n = 1$)				
Phenanthrene	1160			
Anthracene	199			
Fluoranthene	2130			
Pyrene	2570			
Benzo[c]phenanthrene	299			
Benz[a]anthracene	855			
Chrysene	1240			
Benzo[*b*]fluoranthene	885			
Benzo[*k*]fluoranthene	428			
Benzo[*e*]pyrene	828			
Benzo[*a*]pyrene	739			
Perylene	144			
Indeno[1,2,3-*cd*]fluoranthene	54.3			
Indeno[1,2,3-*cd*]pyrene	544			
Dibenz[*a*,*h*]anthracene	82.0			
Picene	92.1			
Benzo[*ghi*]perylene	1220			
Coronene	787			
$\sum$PAH18	14,200			

cordance with the current European filter measurement standard for comfort air filters (Standard EN 779:2012; CEN 2012). This standard defines filter classes using the collection efficiencies of spherical particles with a diameter of 0.4 μm.

During the test with the urban aerosol, the pressure drop of the test objects did not change significantly, and therefore, the initial collection efficiency data were representative for the efficiency during the sampling. All particle collection efficiencies measurements at 0.4 μm was within $\pm 2\%$ for all test objects. Particle collection efficiencies of 0.4-μm particles and TSPs are listed in Table 1.

In the present study, the collection efficiency of only the filter material has been investigated. In real applications, air filters are constructed as filter cartridges (or other constructions) and can contain different amounts of filter material. This does not significantly affect the particle collection efficiency but the airflow resistance. The filter lifetime is dependent on the air filter design and the amount of collected particles (dust) during operation.

To investigate the operational and maintenance performance for air filters (lifecycle cost), it is important to test a filter for a sufficient time period in operation and is not covered in this present study.

Table 3. Reduction efficiency of the mutagenicity +S9 (metabolic activated) and -S9 of the DMSO-soluble fraction of the test objects (mean ± one standard deviation).

	Reduction efficiency of mutagenicity +S9, $n = 3$ (%)	Reduction efficiency of mutagenicity –S9, $n = 3$ (%)
F9	75.4 ± 16.5	81.2 ± 8.9
F7	52.5 ± 9.6[a]	58.5 ± 3.1[b]
City-Flo F7	51.9 ± 32.1[b]	38.2[c]
M6	27.6[c,d]	ns[e]

[a]One measurement at Hornsgatan sampling site included in data.
[b]$n = 2$.
[c]$n = 1$.
[d]Two samples showed no significant reduction in mutagenicity downstream of the test object.
[e]ns = reduction of mutagenicity not significant downstream of the test object.

Removal efficiencies of PAHs

During the sampling campaign at Midsommarkransens gymnasium, the outdoor B[*a*]P concentration of unfiltered air varied between 49.9 and 297 pg m^{-3} (median: 170 pg m^{-3}), similarly the sum of the analyzed PAHs ($\sum$PAH18) concentration varied between 1030 and 4140 pg m^{-3} (median: 2320 pg m^{-3}). The one sample collected at Hornsgatan measured an air concentration of 739 pg m^{-3} B[*a*]P and 14,200 pg m^{-3} of $\sum$PAH18. At that particular sampling occasion, F7 was being used as the test object, and although the PAH concentration measured more than six times higher at Hornsgatan, the removal efficiency was merely about 10% higher than both of the measurements at Midsommarkransens gymnasium, suggesting a consistent performance of the test object at different concentration and composition of the air pollution. The measured air concentrations of individual PAHs at the two sampling sites are summarized in Table 2.

The measured removal efficiencies of $\sum$PAH18 in percent of the different test objects are shown in Table 2. A one-way ANOVA was conducted to compare the removal efficiencies of $\sum$PAH18 of the different test objects. A significant difference between the test objects was found at $\alpha = 0.05$; $F(3, 7) = 27.7211$, $p = 0.0003$. A post hoc Tukey honest significant difference test showed a significant difference between the mean removal efficiency for test object M6 and each of the other tested test objects at a 95% confidence level. No significant difference in removal efficiencies was found between the test objects, F9, City-Flo F7, and F7. Although City-Flo F7 is a combination type filter, which also has the capability of removing gas phase PAHs, there was no significant difference between the test objects in the removal of particles associated PAHs from the otherwise similar F7 filter material. This suggests that any removal of gas phase PAHs does not significantly influence the removal of particle associated PAHs. However, the lower gas phase concentration of the semi-volatile PAHs downstream of the test object if City-Flo F7 is expected to cause an outgassing of semi-volatile PAHs from the particles as the gas phase/particle system reaches equilibrium. However, equilibrium between the gas and particle phase is most likely never established during the air sampling because of the varying concentrations of semi-volatile PAHs in the air.

Reduction of mutagenicity

The mutagenicity of the samples are given in all cases except one, as the linear dose response curves with correlation coefficients (R^2) $\geq$0.9 (Figure 3), indicating that there was little or no toxicity in these concentration ranges used. The mutagenicity per cubic meter air was dependent on the concentration of mutagenic substances in the air during the sampling time and

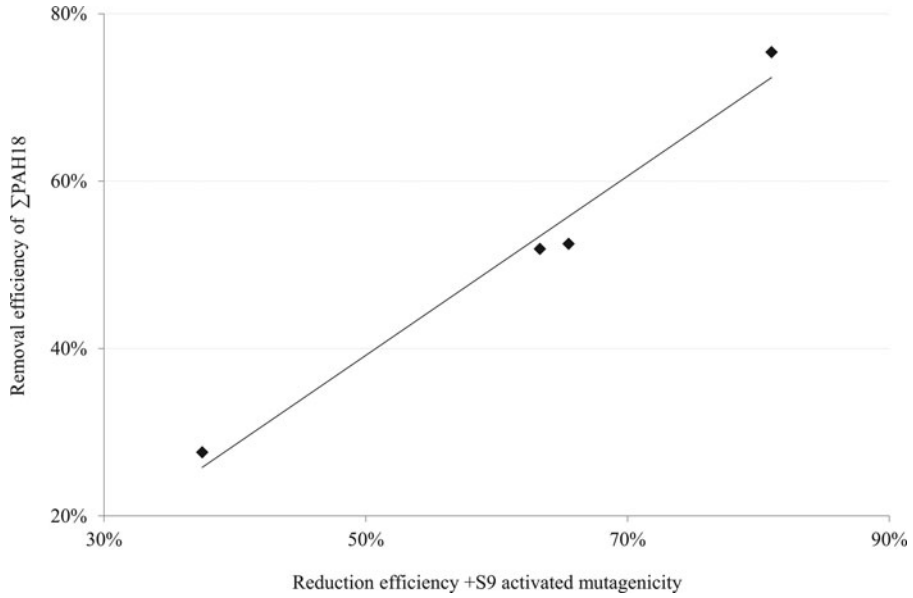

Fig. 4. Removal efficiencies of $\sum$PAH18 versus the reduction of S9 activated mutagenicity (+S9) for the different test objects.

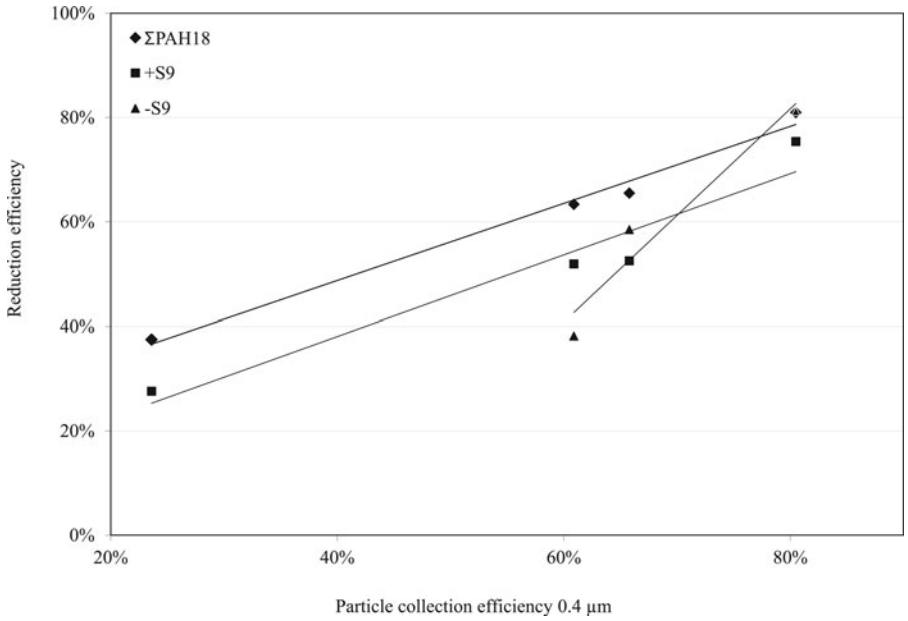

Fig. 5. Removal efficiency of ∑PAH18 and reduction of mutagenicity (both +S9 and –S9) downstream of the test objects versus the collection efficiency of 0.4-μm particles.

was, as expected, found to vary between sampling occasions. Results show that air filter materials with the highest particle collection efficiency also had the highest reduction of revertants per cubic meter of sampled air (Figure 3). The reduction of mutagenicity for the different test objects are summarized in Table 3.

Since PAHs are the major particle associated components contributing to the metabolism-dependent mutagenicity, it is expected that the reduction in mutagenicity in the presence of a metabolizing system (+S9) correlates strongly (R^2 = 0.978) with the removal of PAHs (Figure 4) and also with the collection efficiencies of 0.4-μm particles (R^2 = 0.933); see Figure 5. The results from the data set in Figure 5 indicate that to achieve 50% removal of PAHs from the air stream, there has to be a reduction of at least 45% on 0.4-μm particles. To achieve a reduction of 50% or more in genotoxicity (+S9), there has to be a collection efficiency of at least 50% in 0.4-μm particle size. The corresponding value for –S9 is that there has to be a collection efficiency of at least 60%.

PAH concentrations varied between the different tests with the same type of test object since they were sampled on different occasions. Air also contains other toxic and mutagenic compounds that have not been chemically identified in this present study but are contributing to the mutagenicity of the samples. The measured mutagenicity is the overall effect from all the substances, including possible synergistic and antagonistic effects specific for that particular chemical profile, and can serve as a general assessment of the genotoxicity. However, determining the mutagenic contribution of an individual component in such a mixture, based on its mutagenic property in the absence of other substances, might be problematic since its contribution could be unique for each mixture composition, and such a study would thus be of limited value.

Discussion

By filtering the air supplied from a mechanical ventilation system, the collection of potentially hazardous particles, removal of PAHs and other mutagenic compounds associated with particles in the air stream is achieved. Based on previously published studies showing a lowering of particles in the indoor environment by air filtration, results suggest that the PAH concentration and the genotoxicity of the air in an indoor environment could be lowered by means of air filtration since outdoor particles have PAHs and other genotoxic compounds associated with them. Lowering the indoor particle concentration would result in a lowered indoor PAH concertation for several particle bound PAH isomers. Results show that all of the tested air filters have the capability of removing PAHs from a real urban aerosol and that the magnitude of PAH removal is strongly correlated with the particle collection efficiency. However, Noh and Hwang (2010) concluded from a mass balance model that a minimum filter performance corresponding to MERV11 (corresponding roughly to a M6 classification according to Standard EN779:2012 [CEN 2012]) was required for a housing unit with a size of 150–300 m^3 to sufficiently reduce indoor particle concentrations. However, the present data suggest that a M6 class filter does not significantly reduce directly acting mutagens (–S9), which typically include nitro substituted PAHs. Furthermore, two of the samples from this test object showed no significant reduction of metabolism-dependent mutagenicity (+S9). Thus, the selection of an air filter could have a significance impact, not only on the indoor particle concentration but also on the chemical composition and mutagenicity of the indoor air; however, this needs further investigation.

The possible consequences of air filtration on PAH composition and mutagenicity of the indoor particles in a

real-life setting is difficult to predict from data obtained in this present study given the many factors that influence the indoor air quality besides filter performance, such as air exchange rate, indoor sources of PAHs and other hazardous compounds, indoor activities, the characteristics of the ambient outdoor pollution, infiltration rates, etc. Furthermore, there is a multitude of sources contributing to particle pollution and the composition of PM is diverse, making it hard to point out a specific cause for adverse health effects from PM inhalation. This gap of knowledge makes the feasibility of accurately predicting the possible health benefit from air filtration on in real life solely based on the Ames test and PAH measurements very limited if not impossible. Nevertheless, the present data suggest that is it possible to predict the PAH removal and reduction of genotoxicity in the air supplied to the interior from the outdoor based on the standardized filter classification. The data would be possible to use in mass balance modeling of an indoor environment together with cancer risk assessment using data derived from epidemiological studies of PAH exposure to evaluate the possible lowered risk of cancer due to a reduced exposure to PAHs.

Conclusions

All of the tested air filter materials remove PAHs from a real urban aerosol. However, a significant difference in the PAH removal efficiency was observed between the air filter material with the lowest particle collection efficiency and the other tested air filter materials. There is a possibility of reducing genotoxic effects from outdoor pollutants by using air filters. However, the class/efficiency of the air filter is an important factor. The filter material with the lowest performance did not significantly reduce direct acting mutagens downstream of the air filter, which is indicative that the filter lacks the capability of removing certain mutagenic chemical compounds not identified in this present study.

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