Be Aware of the Indoor Air. Physicochemical Characterization of Airborne Fine Particles in Occupied Homes

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Omelekhina, Yuliya

2022

Document Version:
Publisher's PDF, also known as Version of record

Link to publication

Citation for published version (APA):
Omelekhina, Y. (2022). *Be Aware of the Indoor Air. Physicochemical Characterization of Airborne Fine Particles in Occupied Homes: Physicochemical Characterization of Airborne Fine Particles in Occupied Homes*. Ergonomics and Aerosol Technology, Department of Design Sciences, Lund University.

Total number of authors:
1

Be Aware of the Indoor Air

Physicochemical Characterization of Airborne Fine Particles in Occupied Homes

Yuliya Omelekhina

DOCTORAL DISSERTATION

by due permission of the Faculty of Engineering, Lund University, Sweden.
To be defended at Stora Hörsalen, IKDC. Date 4th of February 2022. Time 9:00.

Faculty opponent
Dr. Jakub Ondráček

Institute of Chemical Process Fundamentals

Academy of Sciences

Prague, Czech Republic

Organization
LUND UNIVERSITY

Document name Doctoral Dissertation

Department of Design Sciences
Ergonomics and Aerosol Technology

Date of issue 2022-02-04

Author: Yuliya Omelekhina

Sponsoring organization

Title and subtitle Be Aware of the Indoor Air
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Negative health effects of exposure to particles of outdoor origin have been confirmed by epidemiological studies. In developed countries, we spend on average 65% of our time in our homes. Thus, the properties of airborne particles indoors need to be understood. The aim of this PhD thesis was to investigate the differences in physicochemical and toxicological characteristics of fine particles (PM2.5) inside and outside occupied homes, as well as to understand the contribution of indoor sources to exposure indoors. The effects of energy renovation and of the occupants' activities on indoor concentrations were assessed.

Indoor and outdoor differences in physicochemical and toxicological characteristics of PM2.5 were studied in 15 homes in urban and rural areas of southern Sweden. PM2.5 characterization was performed with online state-of-the-art techniques, simpler portable instruments, and with offline methods. The occupants' self-reported activities were used to identify the contribution of indoor sources and for interpretation of the results. Measurements in homes were supported by a laboratory study focused on the characterization of particle emissions from candles under stressed burning conditions. An in-vivo toxicity study in mice was performed to assess the differences in toxicological properties of PM2.5 collected indoors and outdoors by evaluating inflammation in bronchoalveolar lavage cells. To understand if the energy renovation affect particle concentrations, measurements were performed inside and outside of seven occupied apartments over three consecutive years, before renovation, after renovation and at a follow-up.

High number of ultrafine particles (UFP) were observed mainly due to the presence of indoor sources such as cooking, and candle burning in homes. Some of the indoor sources additionally contributed to elevated PM2.5 and black carbon (eBC) mass concentrations. In one apartment, a detailed online characterization using a mass spectrometric technique showed that PM1 emissions from indoor sources (e.g., cooking, e-cigarette vaping) were dominated by organic matter (86% of the total mass). Positive Matrix Factorization (PMF) source apportionment of the organic particle fraction showed that the largest contributors to indoor PM1 were e-cigarettes (50%), followed by cooking (40%), and outdoor infiltration was a minor contributor (10%). Candle burning, under stressed burn conditions in the laboratory experiments, emitted large amounts of UFP number concentration, PM2.5 mass and eBC mass concentrations. The wax and wick composition influenced emissions of eBC, PM2.5 and particle-phase polycyclic aromatic hydrocarbons (PAHs). In homes, candle burning also contributed to elevated levels of UFP, PM2.5 and eBC mass concentrations.

In 15 homes, the chemical composition of particles indoors and outdoors was different regarding metals, PAHs, organics, inorganics and endotoxins. Higher concentration of metals such as Fe, Cr, Al, Zn and Mg were found in particles collected indoors compared to outdoors. This was most probably due to cooking, candle and incense burning. Indoor particles collected in 15 homes showed higher toxicity compared to those collected outdoors. This was most likely linked to higher levels of metals, polycyclic aromatic hydrocarbons (PAHs) and endotoxins in particles collected in all homes indoors in comparison to outdoors.

After energy renovation, the UFP concentrations did not decrease and the observed concentrations were mainly affected by the occupants' activities. In order to reduce exposure to UFP particles, more stringent building regulations for kitchen extraction hoods should be considered. The indoor PM2.5 mass concentration had decreased at the follow-up. This could be a result of a lower amount of PM2.5 generated from indoor activities at the follow-up compared to before renovation, and of decreased infiltration of outdoor particles due to the renovation.

The knowledge obtained in this thesis can be used for developing appropriate strategies to minimize exposure to particles indoors. A combination of methods is needed to effectively remove particles generated indoors and to prevent outdoor infiltration. Additionally, the data presented here can be included in mapping of real-life indoor concentrations and in development of indoor air quality models for exposure assessment.

Key words: ultrafine particles (UFP), PM2.5, black carbon, indoor sources, occupied homes, energy renovation

Supplementary bibliographical information

Language English

ISSN and key title 1650-9773 Publication 70

ISBN 978-91-8039-123-8 (print)
ISBN 978-91-8039-124-5 (pdf)

Recipient's notes

Number of pages: 65

Price

Signature Date 2022-02-04

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LUND UNIVERSITY

Cover photo by Yuliya Omelekhina

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Department of Design Sciences
Lund University

ISBN 978-91-8039-123-8 (print)
ISBN 978-91-8039-124-5 (pdf)
ISSN 1650-9773 Publication 70

Printed in Sweden by Media-Tryck, Lund University
Lund 2022

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Abstract

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Papers included in this thesis

Paper I. Differences in physicochemical properties and toxicity of particles indoors and outdoors of occupied apartments in Sweden

A. Wierzbicka, Y. Omelekhina, A. T. Saber, E. Bloom, L. Gren, B. Strandberg, J. Pagels, N. R. Jacobsen. Manuscript to be submitted to a scientific journal.

Paper II. Cooking and electronic cigarettes leading to large differences between indoor and outdoor particle composition and concentration measured by aerosol mass spectrometry

Y. Omelekhina, A. Eriksson, F. Canonaco, A. S. H. Prevot, P. Nilsson, C. Isaxon, J. Pagels, A. Wierzbicka. Environ Sci Process Impacts. 2020. 24;22(6):1382-1396. https://doi.org/10.1039/D0EM00061B

Paper III. Effect of energy renovation and occupants' activities on airborne particle concentrations in Swedish rental apartments

Y. Omelekhina, B. Nordquist, G. Alce, H. Caltenco,, P. Wallentén, J. Borell, A. Wierzbicka. Science of the Total Environment. 2022. 806:149995. https://doi.org/10.1016/j.scitotenv.2021.149995

Paper IV. Emissions of Soot, PAHs, UFPs, NOx and Other health relevant compounds from stressed burning of candles in indoor air

C. Andersen, Y. Omelekhina, B. Rasmussen, M. N. Bennekov, S. N. Skov, M. Køcks, K. Wang, B. Strandberg, M. Bilde, M. Glasius, J. Pagels, A. Wierzbicka. Indoor Air. 2021; 31: 2033– 2048. https://doi.org/10.1111/ina.12909

Paper V. Particulate-bound polycyclic aromatic hydrocarbons (PAHs) and their nitro- and oxy- derivative compounds collected inside and outside homes in southern Sweden

B. Strandberg, Y. Omelekhina, M. Klein, A. Krais, A. Wierzbicka. Manuscript was submitted to a scientific journal.

Author's contributions

Paper I - I conducted all measurements, analyzed online data and produced the graphs. I contributed to the writing of the paper.

Paper II - I performed measurements, analyzed and interpreted the data. I obtained funding for training on the statistical analysis tool - Source Finder (SoFi) for Positive Matrix Factorization (PMF) that was necessary for the study. I performed the PMF analysis based on SoFi software on the AMS data. I invited the Swiss co-authors to collaboration. I wrote the majority of paper.

Paper III – I conducted all the measurements. I had a major role in logistics and planning of the post-renovation measurement campaigns. I performed the data analysis, interpretation and wrote the majority of the paper.

Paper IV - I was responsible for the offline sampling during the measurement campaign. I analyzed the online particle number and mass concentration, and prepared the interpretation and description of them. I contributed to the writing of the paper.

Paper V - I conducted all offline sampling during the measurements and contributed to the writing of the paper.

Acknowledgements

This PhD journey has been a period of intense learning for me. I have had the opportunity to participate in a number of measurement campaigns, to acquire a great deal of practical and theoretical knowledge, and to take part in several conferences.

I would like to thank my main supervisor, Aneta Wierzbicka, for her professional guidance, support, and for introducing me to the scientific field and life in Sweden. I am very grateful to my co-supervisor, Axel Eriksson, for scientific discussions and for providing me with tools that improved my learning during my PhD studies. I would also like to thank my co-supervisor, Christina Isaxon, for the valuable input during the interpretation of the results and during writing of this thesis.

I want to thank my colleagues for the friendly atmosphere and all their support throughout this period.

I am very grateful to my parents and grandparents who always believed in me and supported me throughout my educational journey.

Yuliya Omelekhina

February 2022

Popular science summary

How much time do you usually spend at home? On average, it is 65% in developed countries, which is about 16 hours per day. It is known that exposure to airborne particles of outdoor origin can cause adverse health effects. As we are spending most of our time indoors, we are exposed to airborne particles of different kinds than outdoors. Thus, we have to understand where particles come from, what their properties are, and which sources contribute to indoor levels.

But what are the sources of particles in our homes? Particles can often be generated from activities like cooking, burning candles or incense, use of cleaning products, aroma and salt lamps, and smoking. These indoor sources generate particles smaller than 2.5 micrometers in size, these are called fine particles (PM2.5). Particles also infiltrate from outside though the ventilation system, cracks in the building or just through an open window or door. Additionally, new particles can be formed indoors during chemical reactions between particles and gases of both indoor and outdoor origin.

This PhD thesis focused on the characterization of different properties of fine airborne particles in occupied homes and considers contribution of both indoor and outdoor particle sources. It also investigates the influence of energy renovation and occupants' activities on particle concentration indoors. Additionally, this thesis investigates if particles indoors differ in toxicity in comparison to outdoor particles

We measured PM2.5 inside and outside of 15 homes in southern Sweden. We used online instruments to monitor particle concentrations in real-time and particles were also collected on filters. Occupants reported indoor activities in logbooks and we could link these activities to observed peaks in the particle concentration data. The particles collected on filters were analyzed to determine chemical composition of the PM2.5 and the particles were also used for toxicological studies in mice. During our measurements in homes, we found that candle burning was a strong source of particles. Measurements in the laboratory were hence performed to assess the particle emissions from candles under stressed burning conditions. The effects energy renovation and occupants' activities on particle concentrations were assessed before, after renovation and at the follow-up.

Measurements in fifteen occupied homes showed high concentrations of very small particles called ultrafine particles (UFP, <100 nm), of PM2.5 and of soot. Indoor activities such as cooking, e-cigarette vaping, and candle burning were the major contributors to the indoor particle levels of UFPs. PM2.5 and soot were influenced by both indoor sources and infiltration of particles from outdoors.

Chemical composition of particles emitted during cooking (frying, using the oven, deep-frying) and e-cigarette vaping were dominated by organic material. Particles collected indoors had a higher concentration of metals compared to outdoors. These metals might originate from the food itself or from kitchen equipment when it was heated.

Candle burning emitted high concentration of particles. If the candle flame burns steadily, then the wax reacts with oxygen in the air and carbon dioxide, water and inorganic emissions are produced. However, in real indoor environments the candle flame is frequently disturbed by air movements (e.g., by opened door or window or by people moving around) and this will make the flame flicker resulting in incomplete combustion and hence soot formation. We found during measurements in homes and in the laboratory that candles under stressed burning conditions emit a lot of soot. The wax and wick composition is important as it influences emissions of eBC, PM2.5 and particle-phase polycyclic aromatic hydrocarbons (PAHs).

The toxicological studies we did in mice showed that indoor particles collected in 15 homes were more toxic than outdoor particles collected near the homes.

We have assessed the UFP number and PM2.5 mass concentrations before, after renovation and at the follow-up. It was concluded that the energy renovation did not influence concentrations of UFPs and that these concentrations were mainly influenced by occupants' activities. PM2.5 mass concentration were lower at the follow-up. We found out that it was both due to a decrease of the indoor activities at the follow-up as well as to a reduced infiltration of outdoor particles as a result of the renovation. In order to remove particles that people generate in their homes more efficiently there is a need for stricter building regulations regarding kitchen extraction hoods and ventilation flows. The challenge here is to create a good balance between indoor air quality and energy savings.

The obtained knowledge in this thesis can be used in developing strategy to minimize exposure to particles indoors. This can be achieved by developing methods of effective removing of particles generated indoors directly at the source and by improving ventilation systems to reduce outdoor infiltration.

List of abbreviations

ADS - Activity detection systems

AER – air exchange rate

AMS – Aerosol Mass Spectrometer

BC – black carbon

CE – collection efficiency

eBC- equivalent black carbon

EC – elemental carbon

EF – emission factor

FTX - Exhaust and supply air ventilation with heat recovery

HR-ToF-AMS – High-Resolution Time-of-Flight Aerosol Mass Spectrometer

MS – mass spectra

OC – organic carbon

PAHs – Polycyclic aromatic hydrocarbons

PM2.5 – fine particulate matter

PMF – Positive Matrix Factorization

PNC - particle number concentration

rBC – refractory black carbon

SMPS – Scanning Mobility Particle Sizer

SP-AMS – Soot Particle Aerosol Mass Spectrometer

TEFs - Toxic equivalence factors

UFP – ultrafine particle

Background

Throughout our lives, we are exposed to airborne particles of different composition present in indoor air (1). The particles suspended in the air can be inhaled and may be deposited in different parts of the human respiratory tract depending on particle diameter, particle composition and a persons' breathing pattern. A number of negative health effects have been linked to exposure to outdoor airborne particles, including respiratory and cardiovascular diseases, lung cancer and increased mortality (2-4). The presence of airborne particles of different physicochemical composition in indoor air and their possible health effects are still largely unknown. As people spend most of their time indoors (about 65 % in private homes (5-7)), it is important to assess and understand indoor particle properties and concentrations to be able to connect indoor exposure to various health effects (8).

Aim and objectives

The aim of this thesis was to assess the physicochemical and toxicological characteristics of fine particles (PM2.5) in occupied homes. The influence of energy renovation and occupants' activities on indoor concentrations was investigated. This was done to achieve a better understanding of the contribution of different sources to indoor particle levels and particle characteristics to which people are exposed to in homes.

The specific objectives were:

a) To assess the differences in physical, chemical and toxicological particle characteristics inside and outside of occupied apartments (Papers I, II, III, V)
b) To assess the major particle sources indoors (Papers I, II, IV) and their influence on the indoor concentrations (Papers I, II, III, V);
c) To assess the influence of energy renovations and occupants' activities on the ultrafine particle (UFP) number concentration, fine particle (PM2.5) and black carbon (BC) mass concentrations in the occupied apartments (Paper III).

Introduction

Indoor particle concentrations can be affected by the presence of indoor sources, physicochemical processes indoors, outdoor infiltration and other factors such as building characteristics, ventilation type, filtration (if used), and geographical location of the studied indoor environments (1, 9, 10). Other processes that influence indoor particle concentrations are particle deposition on indoor surfaces, resuspension of deposited particles, and chemical reactions leading to secondary aerosol formation (9). In the next subchapters, the processes that affect indoor particle concentrations will be discussed in detail.

Indoor sources of particles

Indoor activities that produce airborne particles and gaseous pollutants include cooking (frying, boiling, baking, toasting, etc.), candle and incense burning, using cleaning products, vacuuming, sweeping, ironing, printing, using salt and aroma oil lamps, air humidifier, vaping e-cigarette and cigarette smoking (11-24).

Cooking activities and candle burning generate high fine (PM2.5, $<2.5\ \mu m$) particle number and mass concentrations (11-15, 17, 19, 20, 25-32). During cooking, degradation of carbohydrates, fats and denaturation of proteins occurs, resulting in formation of airborne particles, gases, and water vapour (10, 19, 20). When organic compounds from cooking products are heated above their boiling point, they evaporate and upon cooling of the hot vapour, particles are formed (23). Most cooking activities such as frying, boiling, baking, toasting emit fine (PM2.5) particles, however, during specific cooking, e.g., sauteing and when spattering happens, particles can be emitted in the coarse mode (33, 34). Number and mass concentrations of fine particles emitted from different types of cooking may vary. Reported (20, 24, 35-39) average particle number concentrations (PNC, in the range of 10-300 nm) from individual activities ranged between $5.7*10^3$ - $1.2*10^6$ cm^{-3} during cooking of breakfast, $2.5*10^2$ - $5.6*10^4$ cm^{-3} during boiling, $1.4*10^4$ -$8.9*10^6$ cm^{-3} during frying, 0.2 $*10^5$ - $2.3*10^5$ cm^{-3} during baking, $1.0*10^5$-$1.6*10^5$ cm^{-3} during toasting, 1.5 $*10^5$ cm^{-3} during stir-frying. Reported average PM2.5 mass concentration from cooking (15, 24, 39, 40) were between 12-389 $\mu g\ m^{-3}$ and varied depending from the meal types. Average PM2.5 mass concentrations varied depending on cooking activity: 30-48 µg m^{-3} during cooking breakfast, 25-389 µg m^{-3} during frying, 13-24 µg m^{-3} during baking, 12-45 µg m^{-3} during toast making, 25 µg m^{-3} during stir-frying in both an experimental house and in homes (15, 24, 39, 40). Studies in both an experimental house (22, 24) and in laboratory settings (25, 41) showed that the chemical composition of PM2.5 emitted during cooking was dominated by primary organics.

Candle burning may produce different amounts of particles in terms of number and mass depending on the burning mode and candle type. When the candle flame was not disturbed during burning (steady burn) in the laboratory conditions particle number concentrations (range of 2.4-1000 nm) varied between $0.5*10^6$-$4.9*10^7$ cm^{-3} (12, 32). However, in homes there are often sudden air movements, caused by people moving or by doors opening, which makes the candle flame flicker (sooting burn). Particle number concentrations during sooting burn in the laboratory conditions varied between 0.3-0-$9*10^6 cm^{-3}$ and PM2.5 mass concentrations varied between 100 - 2000 µg m^{-3} (12). Incense burning also contribute to indoor mass concentrations. One study showed PM1 mass concentrations during burning of incense that varied between 50-350 µg m^{-3} and the chemical composition was dominated by organics (42).

Cleaning activities that involve usage of cleaning products, air fresheners, and polishes increase the indoor particle as well as gaseous emissions. In some cases, lead to secondary organic particle formation through various chemical reactions, for example between terpenes in the cleaning product and ozone (43-46). A study found that usage of cleaning products increased the number concentrations of UFP and average concentrations varied between $1.9*10^5$ -2.8 $*10^5$ cm^{-3} (20). Maximum PM2.5 mass concentrations during usage of the cleaning products varied between 11-38 µg m^{-3}(34). Cleaning such as sweeping, hovering, and dusting tend to emit particles in coarse mode (47).

As indoor sources contribute a lot to indoor concentrations, there is a need for properly operated ventilation systems to reduce personal exposure. A study was done assessing the reduction by the kitchen hood of ultrafine particles (UFP, range of 2-100 nm) emitted during cooking on a gas stove and oven. It was found (48) that during cooking under the kitchen hood, the UFP were reduced by 39% - 98%, depending on the burner position and kitchen hood flow rates. Rim et al. (49) studied ventilation effectiveness of removal of fine and coarse particles (>2.5 µm) in indoor environments, and found that fine particles (1 µm) were removed more efficiently than coarse particles (7 µm) as fine particles, due to their lower inertia, better follow the air streamlines to the kitchen hood.

Behaviour, transport, and fate of particle in indoor environment

Phase change

Several studies have looked at outdoor-to-indoor aerosol transformations with offline and online high-resolved spectrometric techniques in indoor environments (50, 51). It was found that semi-volatile and non-volatile compounds behave differently during infiltration from outdoors. Semi-volatile compounds (e.g., SVOC, ammonium nitrate (NH_4NO_3), ammonium chloride (NH_4Cl) can partition between gas-phase and particle-phase, depending on concentration, temperature (T) and relative humidity (RH). During the colder time of the year, rapid changes of T and RH between cooler outdoor and warmer indoor environments leads to evaporation of particle phase species indoors (50, 52-57). Non-volatile compounds (i.e., ammonium sulphate ($(NH_4)_2SO_4$), BC) are always found in the particle phase. During outdoor-to-indoor transport, the infiltration of those species is not affected by phase changes, and is size dependent.

Infiltration of outdoor particles

Indoor particle concentration is affected by outdoor-to-indoor transport of outdoor origin particles. Sources of fine particles outdoors are many, including e.g. vehicle exhaust, wood burning, forest fires as well as outdoor cooking and sea salt spray (58-61). The chemical compositions of outdoor PM2.5 usually include sulphate, nitrate, ammonium, other inorganic ions, elemental carbon (EC), organic carbon (OC), and metals (62). At different locations, times, and meteorological conditions the relative abundance of different chemical particle components vary (58).

The fraction of particles that passes through any cracks, gaps or holes in the building envelope is called penetration factor, P (63). Penetration is size-dependent with accumulation mode particles (0.5–2 µm) having the highest penetration efficiency (1) since these particles are too large to have a high diffusivity and are too small to be affected by sedimentation or impaction. A study by Mosley et al. (64) showed that penetration efficiency (2-90 %) through a horizontal slit (0.508 mm high, 102 mm deep, and 433 mm wide) for 2 µm particles increased with increasing indoor/outdoor pressure differences, when for 5 µm particles penetration has not influenced much (1-9 %). For particles smaller than 0.1 µm, penetration efficiency dropped rapidly, reaching almost zero in the 0.01 µm range.

Another term is infiltration factor (15, 65) which includes fraction of the outdoor origin particles that passed though ventilation, cracks, gaps or holes in the building envelope and during regular use of the windows and doors.

$$F_{in} = \frac{aP}{a+k} \quad \text{(Eq. 1)}$$

The infiltration factor (F_{in}, Eq. 1) depends on ventilation (a, AER, h^{-1}), deposition rate (k, h^{-1}) and penetration factor (P) (1, 66). The infiltration factor can be used to estimate the contribution of outdoor origin particles to indoor concentrations during times without influence of indoor sources.

Deposition

Particles deposition on indoor surfaces depends on the particle size, the area and characteristics of the surfaces available for deposition. The deposition loss rate coefficient is defined as the number of particles depositing on available surfaces per unit of time air, and is given in units of h^{-1} (9). UFPs deposit by diffusion and have high deposition rates and particles larger than 1 µm deposit mainly by gravitational sedimentation. Particles with diameters of 0.1 to 0.3 µm have the lowest deposition rate (63). The surface area also influences particle deposition, i.e., the larger the available surface area (i.e., furnished rooms), the more particles will deposit. Additionally, deposition rate will increase if the surface is sticky, charged or colder than surrounding air (9). Presence of airflows (caused by e.g., fans) can increase particle transport towards a surface and enhance the deposition (63, 64).

Resuspension

Particles deposited on the surfaces may become resuspended through activities indoors (such as walking, housekeeping) (9). Ferro et al. (67) reported PM2.5 mass emissions related to resuspension in one house due to the following household activities: folding blankets, folding clothes, dry dusting, making a bed, dancing, vacuuming, walking, and sitting on upholstered furniture.

Indoor/outdoor (I/O) relationship

The particle concentration indoors is affected by a number of parameters which are summarized in the mass balance equation (Eq. 2) (65, 68).

$$\frac{dC_{in}}{dt} = \mathrm{aP}C_{out} + \frac{S}{V} - aC_{in} - kC_{in} \quad \text{(Eq. 2)}$$

where:
V - volume of the room (m^3),
t - time,

a - air exchange rate (h^{-1}),
P - particle penetration factor,
k - deposition rate due to diffusion and sedimentation (h^{-1}),
S - strength of indoor sources (particle number h^{-1} or mass h^{-1}),
C_{in} – indoor concentration (particle number m^{-3} or mass m^{-3})
C_{out} – outdoor concentration (particle number m^{-3} or mass m^{-3})

The *indoor/outdoor (I/O) ratio* (Eq. 3) is used to show the difference between indoor and outdoor particle concentrations, which depends on the infiltration factor, outdoor concentrations, indoor emissions, and indoor losses (by filtration or deposition).

$$I/O\ ratio = \frac{C_{in}}{C_{out}} \quad \text{(Eq. 3)}$$

where C_{in} and C_{out} are the indoor and outdoor particle concentration.

I/O ratio (Eq. 3) based on the data without influence of indoor sources e.g., when no one is at home (represent the infiltration of the outdoor origin particles to indoor concentrations. Different studies have reported I/O ratios in the presence and absence of indoor sources. Morawska and Salthammer (9) found PM2.5 I/O ratios in naturally ventilated buildings with no indoor sources and no human activity from 0.54 to 1.08. When indoor emission sources were present, the I/O ratios ranged from 1 to 2.4.

Health effects of particle exposures

Inhaled particles from various sources may deposit in the human airways and the deposition efficiency depends on their size, water uptake, shape, and density. Different particles sizes deposit in various regions of the respiratory system, due to different deposition mechanisms, or are exhaled. Particles which are larger than 10 µm are deposited by impaction in the head airways (includes nose, mouth, pharynx, larynx). Particles between 2-10 µm are deposited in the tracheobronchial region (from trachea to terminal bronchioles) and 0.5–2 µm particles are mainly deposited in the alveolar region due to sedimentation and Brownian diffusion (69). It is known that exposure to outdoor PM2.5 particles causes negative health effects such as respiratory symptoms, cardiovascular diseases and increased mortality (2, 3).

Some of the components of indoor generated particles, like PAHs, metals, BC and organics are considered carcinogenic (70) and may create a potential risk for human health (PAHs, metals, BC) and lead to formation of reactive oxygen species (ROS) (organics).

Studies were done (71-73) assessing exposures to single indoor sources have shown that the exposure can cause negative health effects. Singh & Li (71) attributed a decrease in lung function and presence of urinary PAH metabolites in kitchen workers exposed to PAHs during cooking. Wang et al 2020 (72) assessed ROS production in vitro from different cooking activities and heating of oils and their impact on genetic damage in human bronchial epithelial cells. It was found that during cooking, ROS are produced and the highest ROS concentrations were produced by sunflower and rapeseed oils. Niu et al. (73) performed in vitro assessment of cytotoxicity to particles emitted from incense and environmental tobacco smoke (ETS). Higher correlations with DNA damage markers and inflammation were attributed to exposure to particle-phase PAHs from incense combustion compared to ETS.

The existing studies underline the importance of studying health effects due to exposure to particles from indoor sources as well as investigating complex real-life indoor mixture in which particles of different origins are present. This is especially important from the health perspective as we spend majority of our time indoors.

Indoor air quality and energy conservation

Energy use has increased worldwide and large amount of energy is consumed by buildings. Energy use needs to be reduced to lower carbon emissions to the atmosphere to tackle the climate change. The EU policy for sustainability includes implementing energy-efficient solutions for the existing buildings (Energy Performance Building Directive (EPBD, 2010), Energy Efficiency Directive (EED, 2012)), for new buildings as well as for future buildings' designs. The transformations are focused towards reducing the overall energy consumption and zero energy/carbon emissions buildings (74). When considering energy-efficient solutions in new buildings and in retrofits it is necessary to find a balance between energy savings and acceptable indoor air quality to ensure health and well-being of the occupants.

The main purpose of the ventilation system in a building is to provide suitable indoor air quality by replacing stale indoor air by fresh air. Adequate ventilation should effectively remove generated pollutants to minimize exposure as well as to control the indoor humidity and temperature. Energy in residential buildings is used for heating, ventilation, lighting, domestic and commercial appliances. Heating comprise the largest percentage of the consumed energy in buildings in Scandinavia, but not all generated energy is optimally used. If the building envelope is not well insulated, the heat would be lost. Thus, an energy renovation needs to ensure the energy performance of the building as well as maintaining optimal settings of the ventilation system to provide fresh and pollutant free air indoors (75).

Ethical considerations

During all measurements in the occupied homes participants were volunteers, they were given information about the project, measurements procedure and our contact details. Occupants were asked to log performed activities that can generate particles (cooking, candle/incense burning, cleaning, cigarette, e-cigarette, etc), keep logbook when they are present/absent from home. Participants were informed in advance about any related risks due to noise, disturbance by visits (installation, check-up, removing the instruments). Participants gave their written consent for participation in the study and knew that they can cease their participation at any time. We ensured data protection, privacy and confidentiality in processing and publishing the data.

During the study of physicochemical and toxicological particle characterization in 15 occupied homes, no personal information and no biological samples were collected, and after discussions with the Ethical Review Board in Lund it was decided that this study does not require ethical approval.

The particle measurements during the renovation study was a part of a larger project that included other indoor air quality measurements, questionnaires and interviews with occupants regarding health symptoms and their perception of thermal comfort, air quality, noise environment and daylight. The study has been approved by the Ethical Review Board (Dnr 2016/841) in Lund and follows Lund University's ethical guidelines.

The in vivo studies in paper I were conducted by National Research Centre for the Working Environment (NRCWE) in Denmark. All animal procedures were reviewed and approved by the local Animal Welfare Body at NRCWE in Denmark, and by the Animal Experiments Inspectorate under the Danish Ministry of Justice. Personnel performing animal studies, was approved to carry out intratracheal instillation of particulate matter with wild type mice, as specified in approved application 2015−15−0201−00465.

Methodology

This thesis presents particle measurements in 15 homes and one chamber study. The physicochemical and toxicological characteristics of fine particles (PM2.5) were assessed with online and offline techniques sampling inside and outside occupied homes (Paper I, II and V, Table 1).

In one of the studied apartments (Paper II, Table 1), indoor and outdoor differences in PM1 chemical particle characteristics and outdoor-to-indoor aerosol chemical transformations were investigated in real-time with a High-Resolution Time-of-Flight Aerosol Mass Spectrometer (HR-ToF-AMS) and an Aethalometer. The particle source origins and the major contributors to indoor mass concentrations were identified using Positive Matrix Factorization (PMF) source apportionment.

Among the studied apartments, after the first round of measurements seven of them underwent energy renovations. Ultrafine particle (UFP) number and fine particle (PM2.5) mass concentrations, as well as PM1 black carbon (BC) mass concentrations were assessed and compared before, after renovation and at a follow-up (Paper III, Table 1).

The physical and chemical characteristics of particle and gaseous emissions from five types of pillar candles were investigated under stressed and steady burning conditions in laboratory settings (Paper IV, Table 1). This study was motivated by the fact that during the measurements in homes, large amounts of candle burning events were recorded in the occupants' activity logbooks. Thus, it was important to perform a detailed characterization of candle emissions under controlled laboratory conditions and compare with the measurements in real indoor environments.

Measurement locations

Measurements in homes. Measurements were performed inside and outside of 15 homes in rural and urban areas of Lund and Malmö, Sweden. The homes comprised houses and apartments with volumes between 115-645 m^3 and with different ventilation systems (Table 1). All homes were located at a distance between 100-500 m from a major road.

Online and offline measurements were performed simultaneously during one-week periods inside and outside of the studied homes. Apartment 5 was an exception, with more detailed measurements that lasted 16 days (Paper II). In all homes, particle measurements were performed inside and outside with identical sets of instruments placed in specifically designed cases (Figures and detailed descriptions are provided in Papers I and III). Cases with instruments inside all measured homes were placed in the living room or hall with the exception of two apartments (7, 10) where instruments were placed in the kitchen for practical reasons.

The apartments in Paper III (Table 1) underwent energy renovation (see details in the next section). Particle concentrations in these apartments were measured over three consecutive years, i.e. before, after renovation and at the follow-up. Particles were also corrected for the PAH analysis during the measurements after renovation, however no particles were collected for chemical offline analysis, endotoxins and toxicological studies at the follow-up.
The outdoor measurements were performed outside of the studied houses, and in case of apartments, instruments were placed on the balcony. Due to practical reasons, the instruments were placed on one of the balconies during the measurements in homes 6-15 before, after renovations, and at the follow-up. The placement of the outdoor measuring case was within 300 m of the studied apartments.

Table 1. Summary of the studied homes (Papers I-IV)

Home № Paper I	**Home № Paper II**	**Home № Paper III**	**Apartment/house**	**Floor area, m^2**	**Volume of the homes, m^3**	**AER, h^{-1}**	**Type of ventilation**	**Number of residents**	**Occupancy, %**	**Location and detailes of the measurement site**	**Distance of each location to the main road**
1			House	285	645	0.6	Natural*	3	84	Rural area, Southern Sweden	500 m
2			Apartment	85	212	1.2	FTX***	2	79	Newly built apartment, Malmö	100 m
3			House	250	625	0.4	Natural*	7	76	Lund	300 m
4			House	110	275	0.6	Natural*	4	91		
5	**5**		Apartment	117	322	0.5	Natural*	4	99	Malmö	
6		**7**	Apartment	66	164	0.5	Mechanical**	1	-	Residences in the multi-family buildings in Lund, built in 1970s	
7		**1**	Apartment	66	164	0.6	Mechanical**	2	84		
8		**2**	Apartment	86	215	0.3	Mechanical**	2	85		
9			Apartment	66	164	1.6	Mechanical**	3	77		
10		**3**	Apartment	66	164	0.4	Mechanical**	1	94		
11			Apartment	86	215	0.4	Mechanical**	1	-		
12		**4**	Apartment	87	218	0.3	Mechanical**	4			
13		**5**	Apartment	46	115	0.5	Mechanical**	1	64		
14			Apartment	80	200	0.9	FTX***	4	73	Newly built apartment, Lund	
15		**6**	Apartment	46	115	0.6	Mechanical**	3	94	Residence in the multi-family buildings, Lund	

*Equipped with kitchen hood
**Mechanical exhaust ventilation system
*** Exhaust and supply air ventilation with heat recovery (FTX system)

Energy renovations. The energy renovation focused on reducing heat losses by providing extra insulation that tightened the building envelope; along with replacing curtain wall windows, and balcony doors on the first and second floors on the south façade. Changes were also made in the ventilation system. Small ventilation vents were installed in the living room and bedroom windows instead of wide window-like ones, which reduced cross-section area of the vent. Kitchen hoods were replaced in all the apartments during the renovation. Bathroom exhaust airflows were increased according to the Swedish practice values (76, 77) which in turn increased the air exchange rates (AER, h^{-1}). More detailed information can be found in Paper III, methods section.

Chamber study. Particle and gas emissions were measured from five types of pillar candles in a stainless steel chamber (21.6 m^3) under stressed burning conditions. Details of the candle types are presented in Paper III Table 1. Table 2 lists the instruments and techniques used for quantification and characterization of airborne emissions. During each experiment three candles were burned for 3.5 h with an AER of 2 h^{-1} inside the chamber. Stressed burning conditions were created by a rotating fan that caused the candle flames to flicker. Two replicates were performed for each candle type. One steady burn experiment was performed for candle 1 to compare results with the stressed burn experiments. At steady burn experiment each candle was put inside a metal mesh cylinder to protect the flame from air movements, ensuring stable conditions.

Above the flame, measurements were performed under stressed conditions for each candle type, and one single candle was used in each experiment. Candle emissions were diluted (1:8) with an ejector dilutor (Model DI 100; Dekati; Finland) and measured with an AMS and NO_x chemiluminescence analyzer. Additionally, candle emissions were measured upon extinguishing the candle. More detailed descriptions of the experiments' procedures are presented in Paper IV.

Aerosol measurements

Particle characteristics and measurement techniques used in this thesis are presented in Table 2. A detailed description of each instrument is presented below.

Table 2. Particle properties measured and techniques used in this thesis

Particle characterisitics	**Intruments/analysis**	**Paper I**	**Paper II**	**Paper III**	**Paper IV**	**Paper V**
Online analysis						
Particle number concentration						
PNC (2.4-914 nm)	Scanning Mobility Particle Sizer (SMPS)				x	
UFP (10-300 nm)	NanoTracer	x		x		
Particle mass concentrations						
PM2.5	DustTrack Aerosol Monitor*	x	x	x	x	
PM1 Organic, NO3, NH4, SO4, Chl	HR-TOF Aerosol Mass Spectrometer		x		x	
BC (PM1)	Aethalometer		x		x	
BC (PM0.9)	Micro-Aethalometer	x		x		
Offline analysis						
PM2.5 mass concentrations	Gravimentic analysis*	x	x	x	x	x
Organic and elemental carbon	OC/EC analysis	x			x	
PAHs	High-resolution gas chromatography/low-resolution mass spectrometry (HRGC/LRMS).	x			x	x
Metals	Inductively Coupled Plasma Mass Spectroscopy (ICP-MS)	x				
Endotoxins	Endotoxins - Limulus Amoebocyte Lysate (LAL) assay; GC/MS	x				
Toxicity	Lung inflammation, DNA damage in broncheoalveolar lavage (BAL) cells	x				

* used for Dusttrak

Online instrumets

High Resolution Time-of-Flight Aerosol Mass Spectrometer (Aerodyne Research Inc., USA) were used to measure time-resolved mass concentrations of non-refractory (organics, sulfate, nitrate, ammonium, chloride) particles in one apartment and in the chamber study.

The operation principle of an AMS (Figure 1) is that aerosols are sampled through a 100 µm critical orifice into an aerodynamic lens, where the particles are focused to a tight particle beam. There, the aerosol is passing through a series of apertures to prevent scattering of the small particles caused by Brownian diffusion and losses of larger particles to the walls by impaction. When aerosols exit the lens, the pressure drop accelerates particles into the sizing region and the particle diameter is obtained by measuring particle time of flight. The AMS detects particles in the size range of 50-500 nm. From there, particles are impacted on a heated surface (~600 °C) where non-refractory aerosol components flash vaporize. The resulting vapor molecules are ionized by 70 eV electrons (tungsten filament is used as the electron source). The electrons, colliding with vaporized molecules, pull out the electrons from the molecular electron shells, convert them into positive ions that are analyzed by mass spectrometry according to their mass-to-charge ratio (m/z) (78, 79).

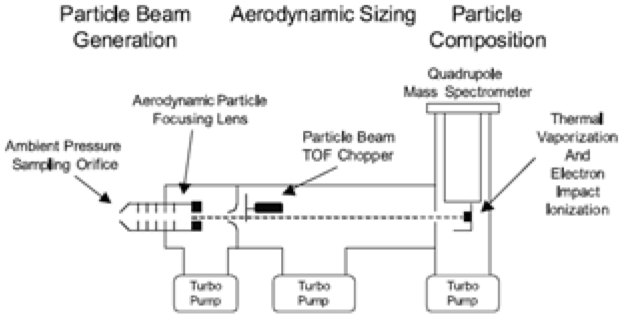


Figure 1. Schematic of an Aerodyne Aerosol Mass Spectrometer (79)

In order to quantify the mass concentration of chemical species, an ionization efficiency calibration of the AMS was performed with 300 nm (mobility diameter) ammonium nitrate and ammonium sulfate particles before and after the measurements. A collection efficiency (CE) of 1 was applied to the indoor and outdoor datasets during apartment measurements for consistency. Particles measured indoors were likely liquid as most of the mass was emitted from cooking and e-cigarette vaping, thus they would have high CE (80, 81). The time resolution of the AMS during measurements inside and outside the apartment was 30-seconds.

The **DustTrak DRX** (8533, TSI; 8520, TSI) was used to estimate real-time PM2.5 mass concentration indoors and outdoors of the apartments. The operation principle of the instrument is that upon illumination, airborne particles in sensing volume scatter light that is detected by a photodetector. The scattered light is converted to voltages which in turn is proportional to the mass concentration. The voltages are converted to mass concentration based on internal calibration (Arizona test dust as reference material). In parallel, particle mass was collected on a filter for gravimetric analysis and the online data were corrected by gravimetric correction factors. The DustTrak that measured inside the homes collected PM2.5 mass for this correction on a filter inside the instrument. Outdoor PM2.5 mass was collected separately on a filter connected to a PM2.5 cyclone and a pump (at 4 l min^{-1}).

An **Aethalometer** (AE33, Magee Scientific) was used to monitor black carbon (eBC) mass concentrations in one apartment and in the chamber study. The instrument principle is that it collects particles on a filter and measures transmission of light through one portion of the filter tape containing the sample, versus the transmission through an unloaded portion of the filter tape (which is a reference area). The attenuation of transmitted light is measured at seven wavelengths. The light absorption is converted to an equivalent BC mass concentration (via mass absorption coefficient (MAC) (82). For current analysis, attenuation at 880 nm wavelength was used. Abbreviation of ‘BC’ instead of ‘eBC’ was used in Paper II.

A **Microaethalometer** (AE51) monitored black carbon (eBC) mass concentrations inside and outside of apartments. A PM2.5 microcyclone was used during sampling at a flow of 150 ml min^{-1}, which corresponded to a cutoff diameter of <900 nm. The operation principle consist of measuring light transmission (at 880 nm) though one spot by a LED light source and a photodiode detector. The absorbance of the spot (attenuation, ATN) is measured relative to a reference portion of the filter. The accumulation of optically-absorbing particles leads to an increase in ATN and optical absorbance is converted to eBC mass concentration via mass absorption coefficient (MAC). Abbreviation of 'BC' instead of 'eBC' was used in Paper III.

Black carbon (BC) mass concentration can be quantified with multiple methods, as BC can be emitted by different combustion processes and have different properties. Optical methods (i.e. Aethalometer, Microaethalometer) that measure light absorption or attenuation and convert it to equivalent black carbon (eBC) applying mass absorption coefficient (MAC) (82). Thermal–optical method measures carbon present in filter samples and quantifies elemental carbon (EC), often closely related to BC, and organic carbon (OC) fractions. Another method is laser-induced incandescence (LII) method that convert thermal emissions to the mass of refractory material present in sampled particles (rBC) (Single Particle Soot Photometer (SP2) (83).

A **Nano Scanning Mobility Particle Sizer** (nano-SMPS, TSI, DMA 3085, CPC model 3776, TSI Inc., USA, measuring a particle size range of 2.4-79.1 nm) and a **long SMPS** (TSI, DMA 3081, CPC model 3775, TSI Inc., USA, measuring a particle size range of 18.8-914 nm) were used to assess particle number concentration and size distributions during the chamber experiments.

In an SMPS, particles are charged in a bipolar charger before they enter the Differential Mobility Particle Sizer (DMA) where they are size selected based on their electrical mobility. After that, the particles enter the Condensation Particle. Counter (CPC) where they pass through a saturated alcohol vapor (butanol) with subsequent cooling in a condenser. The alcohol condenses on particles creating droplets large enough to be detected by optics. To investigate the volatile mass fraction of the UFPs during the candle chamber study, a catalytic stripper (CS) was used in combination with the nano-SMPS. It operated at 350°C and 1.5 l min^{-1} (Model CS 015, Catalytic Instruments GmbH) and was introduced upstream the nano-SMPS during the last 30 minutes of the experiment.

A **NanoTracer** (Oxility Aerasense, Eindhoven, Netherlands) was used to measure particle number concentrations in the size range between 10 and 300 nm in the apartments. The accuracy of the NanoTracer is ±50 % (84). (84)(84)(84)(84)(84)(84)(84)(84)(84)(84)(84)Thus, comparisons were made between the NanoTracers and the long SMPS for the same particle sizes in laboratory conditions prior to and after the measurements. A set of experiments were performed to compare the instruments consisting of background concentrations, low and high concentrations during burning of candles, incense and frying in the 22 m^3 stainless steel chamber. Correction factors were calculated from the comparisons and applied to the data from each of the NanoTracers for each of the measurement campaigns.

Testo sensors (Testo Solutions Division) recorded indoor and outdoor temperatures, (T, °C), and relative humidity (RH, %) during measurements in the apartments.

The **air exchange rates (AERs)** in all the apartments studied were measured with a tracer decay method. N_2O was released in the apartment and dispersed by using two fans. G200 N_2O monitor (Bedfont Scientific Limited) was used to measure the decay of N_2O.

Offline methods

A gravimetric analysis - was performed in order to correct online measurements of PM2.5 mass concentrations measured by the DustTrak. Particles for the gravimetric analysis were collected on the individual filters (37 mm, pore size 2 µm, Teflon Pall Corporation, Port Washington, N.Y.) inside and outside of 15 homes. After gravimetric analysis, the filters were cut into three separate parts and analyzed for PAHs (half a filter), metal content (a quarter of a filter) and inorganic ions (a quarter of a filter), (Papers I, III).

Particle-phase **PAHs** were analyzed in 15 homes (Paper I and V) and particle- and gas- phase PAHs were analyzed during the candle study (Paper IV). For the analysis in 15 homes, a half of each filter (37 mm, pore size 2 µm, Teflon Pall Corporation, Port Washington, N.Y) previously used for gravimetric analysis was analyzed for the following PAHs: naphthalene, 2-methylnaphthalene, 1-methylnaphthalene, biphenyl, 2,3-dimethylnaphthalene, acenaphthylene, acenaphthene, 2,3,5-trimethylnaphthalene, fluorine, 1-methylfluorene, phenanthrene, anthracene, 2-methylphenanthrene, 3-methylphenanthrene, 1-methylphenanthrene, 1-methylanthracene, 2-phenylnaphthalene, fluoranthene, pyrene, 1-methylfluoranthene, 1-methylpyrene, retene, benzo(a)anthracene, chrysene, 2-methylchrysene, benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(a)pyrene, perylene, indeno(1,2,3-c,d)pyrene, dibenzo(a,h)anthracene, benzo(g,h,i)perylene using an Agilent 5975C mass spectrometer (MS) coupled to a 7890A gas chromatograph (GC, Agilent Technologies). The analysis is described in detail in Paper V.

In the candle study, PAHs were sampled with two sampling assemblies. In the first assembly, PM2.5 mass was sampled on a filter (37 mm diameter, 2 µm pore size, Teflon Pall Corporation, flow rate of 2 L min^{-1}). The PM2.5 filters were analyzed for particle PAHs. In the second assembly, a solid adsorbent tube (XAD-II; 120 mg) collected gas-phase PAHs. A filter (the same type as for sampling of PM2.5) was placed in front of the adsorbent tube to remove particles and adsorbent tube was used to quantify gas-phase components. Concentrations of 33 PAHs including 16 US EPA priority PAHs and 16 alkylated species were determined. Toxic equivalence factors (TEFs) of the 12 PAHs were calculated according to Nisbet and LaGoy (85) (Papers I, IV, V). The individual PAH compounds were ranked according to cancer potency relative to BaP (i.e., BaP equivalents), with a TEF factor for BaP set to 1.

An analysis of metal content was performed on the collected PM2.5 mass in 15 homes. A quarter of the individual filters (37 mm, pore size 2 µm, Teflon Pall Corporation, Port Washington, N.Y previously used for gravimetric analysis) was analyzed by inductive coupled plasma mass spectrometry (ICP-MS; iCAP Q, Thermo Fisher Scientific, Bremen, GmbH Germany) (Paper I).

An **inorganic ions analysis** was performed from the collected PM2.5 mass in 15 homes and during the candle study (Papers I, IV). A quarter (in 15 homes) of the individual filters (37 mm, pore size 2 µm, Teflon Pall Corporation, Port Washington, N.Y previously used for gravimetric analysis) and the whole filters (candle study) were analyzed using ion chromatography for detection of Cl, NO_3, SO_4 and PO_4 anions.

Elemental carbon and organic carbon (OC/EC) analysis was performed on the extracted PM2.5 particles from 15 homes and during candle study with a thermal optical analyzer (DRI Model 2001 OC/EC Carbon Analyzer, Atmoslytic Inc., U.S.A.) using the EUSAAR2 protocol (86). OC/EC analysis is the thermal-optical method when the carbonaceous material in aerosol particles is thermally desorbed from a filter, first in an inert atmosphere (He) (OC desorption) and then in an oxidizing atmosphere (He/O2) (EC combustion) at high temperature (86).

Endotoxin analysis was performed on the collected PM2.5 mass inside and outside of 15 homes. PM2.5 was collected on the filters (47 mm, PTFE) using PM2.5 cyclone at flow 15 l min^{-1}. Analysis was performed using kinetic chromogenic Limulus Amoebocyte Lysate (LAL) assay. Individual filters were put in in glass tubes and stored at -20 degrees C. Filters were extracted, diluted and analyzed on an Endosafe® nexgen-PTS™ (Charles River Inc., Wilmington, Massachusetts, USA). Detailed analysis is described in Paper I.

PM2.5 particles for **toxicological studies** were collected with a Dekati Gravimetric Impactor (DGI, 70 l/min, Dekati Ltd, Finland) on 70 mm filters (90 mm Fluoropore PTFE, cut to 70 mm, pore size 3 µm, Merck KGaA, Germany) inside and outside of 15 homes. Extraction and pooling to indoor and outdoor samples is described in detail in paper I. The dry particles were suspended in NanoPure water with 0.1% Tween80. Tween80 is a polyethylene sorbitol ester with stabilizing and emulsifying properties. Carbon Black Printex 90/XE-2B was used as a positive control. Seven weeks old mice (female C57BL/6 mice, N=6) received a single intratracheal instillation of 18, 54 and 162 μg of a specific type of the pooled particle samples (i.e. indoor, outdoor or blanks). Inflammation in broncheoalveolar lavage (BAL) cells were evaluated 1 and 28 days after intratracheal instillation.

Particles collected, extracted and pooled (to indoor and outdoor samples) for toxicological studies were also analyzed for PAHs, metals and endotoxin content. Comparison to chemical composition of particles from individual homes have also performed (details in Paper I).

Ventilation systems in studied homes

The studies were carried out in homes with natural ventilation, mechanical exhaust ventilation and exhaust and supply air ventilation with heat recovery (FTX system) systems.

A **natural draught ventilation system** is typically used in the houses and apartments built in the 1960s in Sweden (87). The temperature difference between outdoor and indoor air causes air movements indoors. In such systems, the outdoor air comes in through vents and gaps in the building envelope and is exhausted through bathroom and kitchen exhaust ventilation ducts that are connected to the chimney on the roof of the building. Such ventilation system is dependent from the weather. The air exchange is higher in winter because the differences in temperature are higher compared to the other seasons. Our measurements in the homes were carried out during winter. In the measured homes with natural ventilation, kitchen hoods (KH) were installed and were switched on by occupants only when cooking. The AERs in homes with natural ventilation in our studies (Papers I, II) were between 0.39-0.56 h^{-1} (Table 3).

A **mechanical exhaust ventilation system** is an improved version of the natural ventilation system and is typical for buildings from 1970s (87). The difference between mechanical exhaust ventilation systems and natural ventilation is that in the mechanical system, the airflows are mechanically forced into the kitchen and bathroom exhausts. In Sweden, the exhaust flows according to the Swedish practice values (76, 77) should be 10 l/s through kitchen hood and 15 l/s through bathroom exhaust. The required airflow when a kitchen hood is used in the forced mode, which can be manually switched on, should equal to a minimum of 25 l/s (76, 77). The AERs of the apartments with mechanical exhaust ventilation in this study were between 0.31-0.64 h^{-1} before renovation, 0.49-0.73 h^{-1} after renovation, and 0.56-0.8 h^{-1} at the follow-up (Table 3).

An **exhaust and supply air ventilation with heat recovery system (FTX system)** is an energy efficient system and is not dependent on the weather (i.e., temperature difference). In Sweden, it was incorporated into the apartments built from 1990s and onwards (87). The outdoor supply air is filtered, heated/cooled, moisture is removed, and then supplied through the bedroom and living room. The exhaust air is mechanically forced in the kitchen and bathroom. Heat is recovered from the exhaust's warm air in a heat exchanger. The AERs in the apartments that had such a system (Paper I) ranged between 0.85-1.18 h^{-1} (Table 3).

Table 3. AERs of studied apartments before, after the renovation and at the follow-up

№	AER, h^{-1}		
	Before	After	The follow up
6	0.50	0.58	0.57
7	0.59	0.41	0.80
8	0.31	0.49	0.57
10	0.40	0.60	0.75
12	0.31	0.55	-
13	0.51	0.73	0.75
15	0.64	0.53	0.56

Monitoring occupants' activities

Self-reporting During all the measurements, indoor activities as well as presence/absence in homes were recorded by occupants' in activity logbooks. Logged indoor activities included cooking, candle and incense burning, smoking, using salt and essential oil lamps, using household products, vacuuming, sweeping, ironing, printing and opening windows.

Activity-based detection system (ADS) was used to monitor cooking activities performed by occupants in the apartments during measurements before, after renovations and at the follow-up. The ADS motion and temperature sensors were mounted under the kitchen hood and logged cooking activities (using the stove and/or the oven). A magnetic sensor was attached to the kitchen hood and was used to detect when occupants pulled it out to put it in the forced air mode.

Data processing

Data analysis included calculating averages, standard deviation (SD), maximum, minimum and indoor to outdoor ratios (I/O) for the data collected in homes. Indoor to outdoor ratios (I/O) were calculated based on the average particle number and mass concentrations and were used to show differences in the particle concentrations. The averages of particle concentrations during occupancy and non-activity periods were also calculated for all homes. An occupancy period was defined as the time when at least one person was at home. The particle concentrations during occupancy times represented the concentrations to which people were exposed to when at home. Particle concentrations during non-activity periods represented the infiltrated particle concentrations from outdoors. In order to calculate the infiltrated particle fraction, the data were analyzed for these periods: 1) When no one was in the apartment during daytime; 2) During nighttime between 00:00-05:00; 3) In both cases if the indoor concentration was influenced by preceding indoor activities, those periods were excluded. Time-series of particle number and mass concentrations were used for the data analysis, evaluation and sources identification together with logbooks in all Papers.

For the data treatment from HR Aerosol Mass Spectrometer (HR-AMS) in one residence (Paper II) and in the chamber study (Paper IV), a high-resolution analysis was carried out in IGOR Pro 6.37 software with SQUIRREL v 1.6P and PIKA v 1.2P (Wavemetrics, Inc., Portland, OR, USA).

A statistical tool, Positive Matrix Factorization (PMF), was used to interpret indoor organic mass fractions acquired from the HR-AMS collected in one apartment (Paper II). PMF (SoFi 6.3 H, Source Finder, Paul Scherer Institute, (PSI) was used to apportion the major contributing sources to indoor concentration and identify unknown indoor sources that were not logged in the activity logs. The PMF utilizes a matrix (X) where columns are the time series of a factor (G), and the rows are factor profiles (F) (i.e., mass spectra), E is a model residual, Eq. 4 (88):

$X = GF + E,$ (Eq. 4)

Emission factors based on the number concentrations were calculated (Eq.5) during candle burning observed in 15 homes (Paper I) and compared with the emission factors obtained during chamber studies.

$EF = C_{av} * V * (a + k)$ (Eq. 5)

Where C_{av} - average concentration (number concentration),

V - home volume (m^3),

a - air exchange rate AER (h^{-1}),

k - decay rate (h^{-1}).

a+k values were calculated from the measured data for each decay of candle burning event.

For the chamber study (Paper IV) emission factors (Eq. 6) were calculated for all five candle types under stressed burning conditions. All emission factors were calculated based on measured particle number and mass concentrations (65, 68). More detailed explanation of the calculations can be found in Paper IV.

$$EF = \frac{C_{av} * V * (a+k)}{n} \quad \text{(Eq. 6)}$$

Where C_{av} - average concentration (number or mass concentration),

V - chamber volume (m^3),

a - air exchange rate AER (h^{-1}),

k - decay rate (h^{-1}),

n - number of candles burned.

Results and discussions

Particle physicochemical characterization inside and outside of the 15 occupied homes

The data presented in this section provides insight into the physicochemical characteristics of the PM2.5 sampled in the 15 occupied homes (Paper I).

UFP number concentration. Table 4 illustrates the average UFP number concentrations inside and outside of the 15 occupied homes during the entire monitoring period. The average UFP number concentrations were three times higher inside the apartments (8800 cm^{-3}, SD 9300 cm^{-3}) than outside (2800 cm^{-3}, SD 1200 cm^{-3}). The higher UFP number concentrations observed indoors were caused by particles emitted from the occupants' indoor activities. Average occupancy time was 73 %, varying between 46.3 % and 100 %, across studied homes. The most typical activities responsible for elevated indoor concentrations were cooking (frying, using the oven, deep-frying), candle and incense burning. The above-mentioned indoor activities were recorded by the occupants the activity logbooks and peaks caused by these activities could hence be identified. The influence of indoor activities on UFP number concentrations can be seen in the time series for different homes (Paper III, Figures S5-10). There was, however, variability in the average UFP number concentrations between the individual apartments 1000 cm^{-3} (SD 500 cm^{-3}) and 38900 cm^{-3} (SD 120800 cm^{-3}) (Table 4). The differences were due to the different number of activities that occupants had performed (Paper III, Figure S5-11).

UFP number concentrations during occupancy times were higher (10900 cm^{-3}, SD 12100 cm^{-3}) compared to the UFP number concentrations averaged over the whole measurement period. Particle concentration data during occupancy times is important from the personal exposure perspective because it represents concentrations to which people are exposed to while at home.

Particles of outdoor origin contributed to 20 % of the UFP concentrations indoors Infiltration from outdoors was calculated using the non-activity periods i.e., the times when no one was at home, during nighttime (00:00-05:00), and the times when concentrations were influenced by preceding indoor activities were excluded. The influence of particle infiltration from outdoors can be observed in Paper III Figures S5-11. When there was no activity, the UFP number concentrations indoors were similar to or lower than outdoor concentrations. Likely sources of outdoor air pollution in these measurements include traffic emissions from several roadways, local sources of biomass burning (in the area of the private houses), and long-range transport.

Table 4. Summary of the UFP number concentrations for total monitoring, occupancy and non-activity periods

№	Total total monitoring period									Occupancy time			Non-activity time		
	Indoors, cm^{-3}				Outdoors, cm^{-3}				I/O ratio	Indoors, cm^{-3}	Outdoors, cm^{-3}	I/O ratio	Indoors, cm^{-3}	Outdoors, cm^{-3}	I/O ratio
	Average	SD	Max	Min	Average	SD	Max	Min		Average			Average		
1	11700	42100	785100	<100	2600	500	4700	200	4.5	12300	1300	9.5	1800	1200	1.5
2	7400	19300	161200	600	3800	1400	18500	700	1.9	8600	4000	2.2	3200	3300	1.0
3	4700	9700	139800	<100	4300	2300	26700	600	1.1	5300	4000	1.3	2000	4300	0.5
4	10900	32400	753900	200	4400	2600	40600	600	2.5	11200	4400	2.5	1900	3500	0.5
5	4300	4500	63000	400	5300	4100	57100	400	0.8	4300	5300	0.8	2200	3800	0.6
6	1000	500	4800	<100	1800	1400	76900	<100	0.6	-			-		
7	8600	27300	211600	<100	3100	1400	28100	800	2.8	9600	3000	3.2	2000	3100	0.6
8	6300	15700	198800	300	2600	1300	9700	300	2.4	6800	2500	2.7	1800	2700	0.7
9	38900	120830	751700	200	1900	900	9100	<100	20.5	49500	1900	26.1	1600	1800	0.9
10	13400	75800	1535600	<100	1300	600	6300	<100	10.3	14200	1300	10.9	1000	1100	0.9
11										-			-		
12	6400	41000	6238900	400	1500	1200	74200	<100	4.3	8200	1600	5.1	1300	1400	0.9
13	5400	19600	389300	200	1400	800	25500	<100	3.9	6200	1400	4.4	1200	1400	0.9
14	2700	3800	59000	200	3100	7500	730500	<100	0.9	2900	2700	1.1	2100	3900	0.5
15	2500	14000	350200	100	3000	15900	765600	400	0.8	2600	3100	0.8	900	1700	0.5
Average	**8800**	**9300**			**2800**	**1200**			**4.1**	**10900**	**2800**	**5.4**	**1800**	**2500**	**0.8**

PM2.5 mass concentration. Average PM2.5 mass concentrations indoors in the studied homes (Table 5) were similar (7.5 µg m^{-3}, SD 6.0 µg m^{-3}) to outdoor values (7.3 µg m^{-3}, SD 5.0 µg m^{-3}) during the total monitoring period. A slightly higher average I/O ratio of the PM2.5 mass was observed during occupancy time compared to the total monitoring period due to the influence of the indoor sources (candle burning, cooking).

Table 5. Summary of the PM2.5 mass concentrations for total monitoring, occupancy and non-activity periods

№	Total total monitoring period									Occupancy time			Non-activity time		
	Indoors, µg m^{-3}				Outdoors, µg m^{-3}				I/O ratio	Indoors, µg m^{-3}	Outdoors, µg m^{-3}	I/O ratio	Indoors, µg m^{-3}	Outdoors, µg m^{-3}	I/O ratio
	Average	SD	Max	Min	Average	SD	Max	Min		Average			Average		
1	3.5	10.6	492.7	2.1	5.0	1.1	7.0	0.5	0.7	-			-		
2	3.2	2.2	9.2	0.5	5.3	3.3	74.7	0.4	0.6	3.2	5.6	0.6	1.0	4.6	0.2
3	3.7	4.5	143.1	0.4	5.3	6.2	30.3	0.6	0.7	4.5	4.9	0.9	2.0	6.5	0.3
4	5.2	6.2	65.0	0.7	4.4	5.0	72.4	0.5	1.2	5.2	4.4	1.2	1.7	4.1	0.4
5	6.0	13.4	116.5	0.4	6.8	6.0	30.2	0.4	0.9	6.0	6.8	0.9	2.2	6.8	0.3
6	2.3	2.3	12.8	0.6	4.0	3.6	68.2	1.1	0.6	-			-		
7	11.7	12.1	280.6	0.4	15.1	9.0	50.5	3.1	0.8	11.7	15.1	0.8	12.3	17.3	0.7
8	15.6	12.7	84.0	0.4	21.9	15.6	62.2	3.2	0.7	14.3	21.2	0.7	12.6	22.4	0.6
9	10.8	28.2	1481.9	0.7	10.2	10.0	49.6	0.4	1.1	11.0	10.2	1.1	10.0	10.2	1.0
10	5.5	20.8	1884.8	0.8	6.5	3.9	22.9	1.5	0.8	5.6	6.5	0.9	2.4	5.9	0.4
11	7.5	-			4.7	-			1.6	-			-		
12	5.2	3.6	34.9	1.4	5.5	4.7	18.5	1.2	0.9	7.3	5.9	1.2	5.2	5.3	1.0
13	24.3	189.2	4572.8	0.4	3.7	3.7	109.7	0.6	6.6	24.3	3.7	6.6	4.0	3.8	1.1
14	2.1	1.2	8.5	0.8	5.0	3.7	135.0	1.1	0.4	2.3	5	0.5	2.0	5.0	0.4
15	5.5	5.2	64.7	1.0	6.6	20.2	964.8	1.3	0.8	5.7	6.6	0.9	1.5	4.8	0.3
Average	**7.5**	**6.0**			**7.3**	**5.0**			**1.2**	**8.4**	**8.0**	**1.3**	**4.7**	**8.1**	**0.6**

However, indoor sources were not the main contributors to the average indoor PM2.5 mass and, as can be seen from the non-activity period, the majority of the PM2.5 mass infiltrated from outdoors (60 %).

eBC mass concentration. The average mass concentrations of equivalent black carbon (eBC) were slightly lower indoors (0.4 µg m^{-3}, SD 0.3 µg m^{-3}) compared to outdoors (0.5, SD 0.3 µg m^{-3}) values during the total monitoring period (Table 6). eBC mass concentrations originated from both outdoor and indoor sources (indoor sources were candle and incense burning, frying, and occasional smoking) which is visible from the time series (Paper III, Figures S4-10).

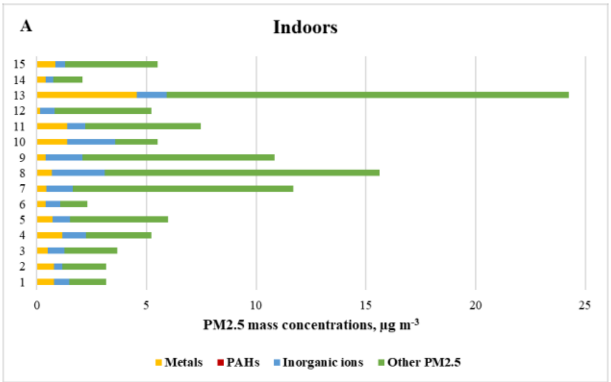
Table 6. Summary of the eBC mass concentrations for total monitoring period

№	Indoors				Outdoors				I/O ratio
	Average	**SD**	**Max**	**Min**	**Average**	**SD**	**Max**	**Min**	
1	0.3	1.0	19.5	0.1	0.2	0.3	2.4	0.1	1.2
2	0.2	0.2	2.2	0.1	0.3	0.2	3.7	0.1	0.7
3	0.3	0.3	2.1	0.1	0.4	0.5	2.9	0.1	0.7
4	0.4	0.3	5.7	0.1	0.3	0.4	4.8	0.1	1.2
5	0.4	0.3	2.8	0.1	0.5	0.4	3.3	0.1	0.7
6	0.2	0.1	1.9	0.1	0.3	0.2	1.0	0.1	0.6
7	0.5	0.5	12.6	0.1	0.8	0.6	3.9	0.1	0.6
8	0.7	1.7	139.8	0.1	1.2	0.9	4.1	0.1	0.6
9	1.2	4.3	81.5	0.1	0.6	0.5	4.7	0.1	1.8
10	0.3	1.1	21.2	0.1	0.3	0.2	2.4	0.1	1.2
11	0.2	0.2	4.0	0.1	0.3	0.1	0.8	0.1	0.8
12	-				-				
13	-				-				
14	0.5	0.3	2.5	0.1	0.4	0.9	111.2	0.1	1.3
15	0.3	0.2	1.9	0.1	0.9	8.2	775.0	0.1	0.3
Average	**0.4**	**0.3**			**0.5**	**0.3**			**0.9**

Offline particle chemical characterization

Figure 2 illustrates the metals, PAH's, inorganic ions and other constituents of the PM2.5 mass concentration indoors and outdoors. The total average concentration of metals in PM2.5 was twice higher indoors (1.0 µg m^{-3}, SD 1.1) compared to outdoors (0.5 µg m^{-3}, SD 0.5) in all homes, as illustrated in Figure 3. The dominating metals indoors were Fe, Cr, Al Zn and Mg. PM2.5 mass concentrations of Fe varied between 0.06 and 2.63 µg m^{-3}, Cr between 0.06 and 0.49 µg m^{-3}, Al between 0.01 and 0.09 µg m^{-3}, Zn between 0.08 and 36.8 µg m^{-3} and Mg between 0.007 and 0.05 µg m^{-3}. These metals originated partly from outdoors but must also have been emitted by indoor sources. Such metals could be emitted from activities such as cooking, candle burning and incense burning. See and Balasubramanian (89) previously reported that metals such as Zn, Fe, Cu, Mn, Pb, Ni, Al, V, Cr were emitted during Chinese cooking. The metals can be emitted from the food but also from kitchen equipment when subjected to high temperatures. For instance, Cu, Fe, Mn, and Zn can be emitted from vegetables (90), Cu, Fe, Zn from meat (91), and Cu, Zn, and Cd from fish (92). The metal concentrations in the 15 homes were generally low. However, if cooking activities are frequent and occur three or more times per day, there is a health concern regarding personal exposure to increased emissions of metals. One of the dominant metals emitted from cooking was Fe, which is known to play a role in the generation of reactive oxygen species (ROS) which, in turn, can cause damage to DNA and induce deleterious health effects (93-95). Additionally, metals such as Cu, Fe, and Zn can accumulate in the body and become a health hazard after chronic exposure even to low levels (96) .

Some specific indoor sources such as vaping e-cigarette (which occurred in home 5), cigarette smoking (which occurred in homes 8 and 11) could have emitted Fe, Al, Cr, As, Cd, Ni, Pb, Zn and Co (97) . Emissions of the above mentioned metals during e-cigarette vaping have been previously reported by Williams et al. (98) and Saffari et al. (99) who suggested that the material used in the cartridge of the e-cigarette is a likely source of those metals rather than the e-liquid.



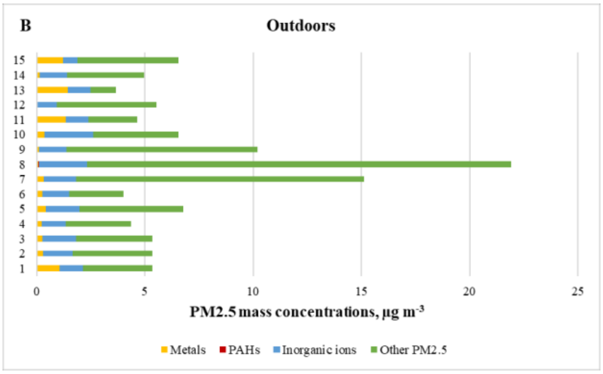


Figure 2. Concentration of metals, PAH's, inorganic ions, and other constituents in PM2.5 indoors (A) and outdoors (B) in 15 homes. Other PM2.5 denotes unknown particle mass.

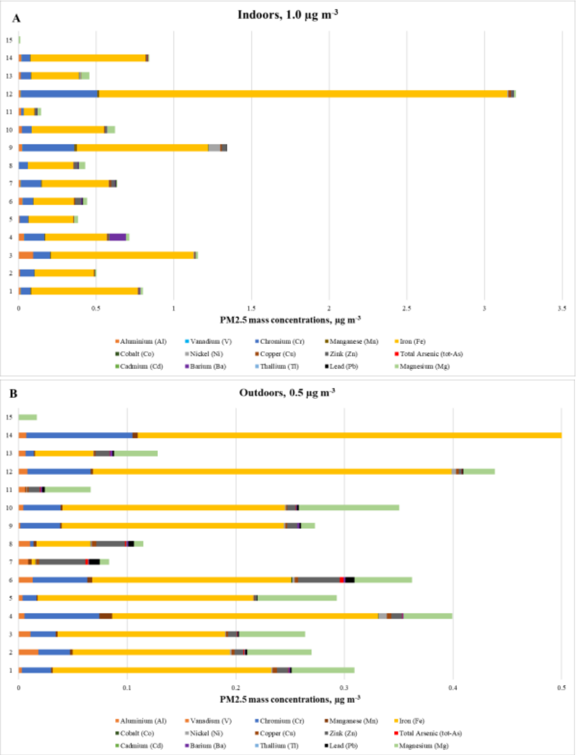


Figure 3. Concentration of specific metals in PM2.5 mass indoors (A) and outdoors (B) in 15 homes

The average concentration of *inorganics* in the PM2.5 was lower indoors (1.0 µg m^{-3}, SD 0.6) than outdoors (1.3 µg m^{-3}, SD 0.4) (Figure 2). However, there was one exception, concentrations of P were higher in all homes. P could partly originate from outdoors and be emitted from indoor sources. In some homes, the indoor concentrations of Ca, K, Na, Mg, Cl were higher compared to outdoors. These these compounds may have been emitted as inorganic ions during candle burning or, cooking activities. Pagels et al. (12) have reported emissions of PO_4^{3-}, K^+, Na^+, Cl^- during burning of stearin-based candle and combinations of stearin and paraffin-based candle in a laboratory setting.

Generally, the average levels of *PAHs* in the PM2.5 were low both indoors and outdoors with a slightly higher average PAH concentration outdoors (0.003 µg m^{-3}, SD 0.006) compared to indoors (0.001 µg m^{-3}, SD 0.001). In home 15, the concentration of PAHs was higher indoors compared to outdoors. PAHs can originate from outdoors or be emitted from indoor sources. PAHs can be generated during combustion processes such as candle or incense burning, cooking (i.e., frying, deep-frying) (Paper IV, V).

The average *endotoxin* concentration was higher indoors (0.27 ng m^{-3}) than outdoors (0.08 ng m^{-3}). Typical sources of endotoxins indoors are pets, humidifiers, pests and outdoor infiltration (100). However, the concentrations of *endotoxins* were generally low in 15 homes. A health-based occupational exposure limit value for endotoxins is 9 ng m^{-3} (average during 8-h), recommended by the Dutch Expert Committee on Occupational Safety (DECOS).

In-vivo toxicological assessment. Inflammation in broncheoalveolar lavage (BAL) cells was evaluated 1 and 28 days after intratracheal instillation. The lung inflammatory response was characterized by an increase in neutrophils and eosinophils cells count after a single intratracheal instillation of 18, 54 and 162 µg of indoor, outdoor and Printex 90 particle samples (Paper I, Figures 7, 8).

Results showed that Neutrophil cells count (Paper I in Figure 7) was seven times higher for indoor particles compared to outdoor particles after instillation of 162 µg after the day 1. Neutrophil cells count was also higher due to indoor particles compared to outdoor ones 28 days after instillation. However, the cells count was ten times lower, both for indoor and outdoor particles after 28 days compared to the first day after instillation. After instillation of the same dose (162 µg) of Printex 90 there was higher Neutrophil cells count compared to indoor particles on the 1st and 28th days. Printex 90 was used as a positive control and is known for its's toxicological effects, so it should cause negative effects.

The levels of Eosinophils count were 7 times higher after day 28 compared to day 1 after instillation of 162 µg of indoor particles. The response to indoor particles was higher compared to outdoor ones and to Printex 90.

Higher toxicological response to indoor particles compared to outdoor ones was observed both after the 1st and 28th day. The effects from indoor particles have decreased after 28 days based on the neutrophil cells count, however, it increased in the case of eosinophils. The levels of eosinophils after instillation of indoor particles were higher compared to after instillation of outdoor particles and Printex 90. Higher toxicity of indoor particles indicates a potential risk for human health as people are exposed to that particle composition on a daily basis and no time is given to recover from those particle types.

Results from the extracted particles also showed that metals and endotoxins concentrations were higher indoors compared to outdoors. The dominating metals in the indoor extracted particles were Fe, Al, Zn and Mg. In comparison to the individual filters, PAHs concentrations in extracted particles used for toxicological assessment showed to be higher in indoor particles. Higher PAH content in the indoor extracted particles could be explained by the lower temperature during sampling indoors for toxicological studies (i.e. outside the sampling case at ambient temperatures) compared to all other samples (inside the sampling case at elevated temperatures, more details in Paper I). Lower temperature (in case of indoor sampling for toxicological study) most probably reduced evaporation and preserved PAHs in particle phase. Higher toxicity of the indoor extracted particles (used for toxicological assessment) was associated with the higher content of the metals, PAHs and endotoxins in comparison to outdoor particles.

Detailed physicochemical characterization inside and outside the occupied residence

Based on the detailed chemical composition measurements in apartment 5 (Table 3, 4, and 5) with use of AMS and aethalometer, the 16-days average particle mass concentration inside the apartment (15 µg m^{-3}, SD 196 µg m^{-3}) was found to be higher compared to the outside (7 µg m^{-3}, SD 6.2 µg m^{-3}). As the apartment was occupied and people were following their regular activities, indoor concentrations were affected by the particle mass emitted from indoor sources. The most typical activities that caused the elevated indoor concentrations were cooking (frying, using the oven, deep-frying), e-cigarette vaping and candle burning.

Indoor sources were mainly responsible for the emissions of organic matter PM1 mass (86% of the total mass, Figure 4). Other indoor particle mass concentrations comprised of black carbon (6%), sulfate (4%) and nitrate (2%).

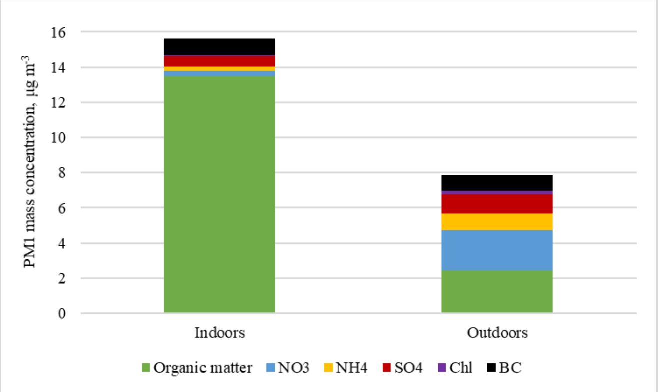


Figure 4. Average chemical composition of particle mass concentration PM1 indoors (left) and outdoors (right) of apartment 5 during the entire measurement period. Presented data are based on HR-ToF Aerosol Mass Spectrometer and Aethalometer measuremnets

The largest fraction of the indoor PM1 mass was organic matter and it was characterized in detail. Positive Matrix Factorization (PMF) was applied to identify how much of the PM1 mass was emitted from each indoor source and to estimate the infiltrated particle mass fraction. With PMF we could identify three indoor organic aerosol (OA) factors: cooking OAI (COAI), cooking OAII (COAII), electronic cigarette OA (EOA); and one outdoor factor infiltrated from outside: outdoor contribution (OOA) factor (Paper II, Figure 6).

On average, the majority of the PM1 mass was emitted from e-cigarette (6.8 µg m^{-3}, 44%, Paper II, Figure 6B). As can be seen from the time series (Paper II, Figure

6B), e-cigarette vaping has a tendency to emit short-lived peaks of high PM1 mass concentrations, and the maximum concentration reached 15 700 µg m^{-3}. However, the e-cigarette was vaped in close proximity to the sampling inlet, thus, the observed concentrations may not be representative for the entire residence. The organic mass spectrum of the e-cigarette OA profile (EOA, Paper II, Figure 5B) showed emissions of glycerine with characteristic peaks at mass-to-charge ratio (m/z) 43 and m/z 61. Glycerine is a main constituent of the e-liquid that is added as humectant for vapor production (101). All e-cigarette events (n=14) were identified by tracing these peaks. The organic mass spectrum found during the measurements in apartment 5 were similar to the mass spectrum of the e-cigarette vaped in laboratory conditions (Paper II, Figure 5C).

Cooking activities (n=29) contributed to 33% (5.0 µg m^{-3}) to the total organic PM1 mass fraction. Two cooking factors, COAI and COAII, were represented by m/z 43, 55, 57, 60, 71 and 73 (Paper II, Figure 6). The ion peaks observed originated from different processes and chemical reactions such as denaturation of proteins, and the degradation of carbohydrates and fats at different temperatures that occur during food treatment. The ion peaks (Paper II, Figure 5B) at C_3H_5 (m/z 41) and C_4H_7 (m/z 55) could originate from the formation of unbranched-chain saturated hydrocarbons and long-chain fatty acids during frying and baking of foods of animal origin (26, 102-105). M/z 29 (CHO^+) appeared either as a result of thermal decomposition of cooking oils (106) or during hydrolysis of fats. Peaks at m/z 60 ($C_2H_4O_2$) and m/z 73 ($C_3H_5O_2$) originated from the fragmentation of sugars during cooking (26, 103). Carbohydrates (e.g., starch, sugars and cellulose) may decompose to single ring sugars though the hydrolysis when heated with water. During the hydrolysis, water reacts with the oxygen atom that joins the sugar rings and breaks complex sugars down into single ring sugars (107). These monomer sugars can decompose to anhydrous sugars such as levoglucosan and similar molecules upon heating. M/z 44 CO_2 release was a result of the thermal decarboxylation of organic acids during cooking (108).

The COAI and COAII cooking factor profiles were different in their intensity of m/z's 60 and 73. The above-mentioned peaks are usually markers of the anhydrous sugars (e.g., levoglucosan) formed during low-temperature pyrolysis of cellulose during biomass burning outdoors. Anhydrous sugars were also detected during cooking (91) with a higher abundance in Chinese cooking style and a lower abundance during meat cooking. COAI may represent low temperature cooking and cooking of food with low carbohydrate content. COA II may represent high temperature cooking of food rich in carbohydrates.

PM1 mass concentrations from candles were not retrieved by PMF because candle burning emits mostly BC, depending on the burning conditions, and salts particles (phosphates and alkali nitrates). Thus, as organic mass concentrations were not emitted or emitted in limited amounts during candle burning, this source could not be identified by PMF. The candle study (Paper IV) confirmed that particle mass emissions from candles burning under stressed conditions were dominated by soot (black carbon, BC) and organics were detected predominantly upon extinction.

The chemical composition outdoors was different compared to the indoors, and was dominated by organic matter (31%) and nitrate (29%), BC (11%), sulfate (14%), ammonium (12%) and chloride (3%). The contribution of the particle mass from outdoors was 1.6 µg m^{-3}. The dominating ion in the outdoor OA mass spectrum was CO_2^+ ion at m/z 44 (Paper II, Figure S4) which appeared as a result of decomposition and fragmentation of oxygenated organic acids, as reported earlier (90,92,103, 105) The typical PM2.5 mass concentrations outdoors (in several monitoring stations) varies between 1-12 µg m^{-3} in the urban and rural areas of southern Sweden (109).

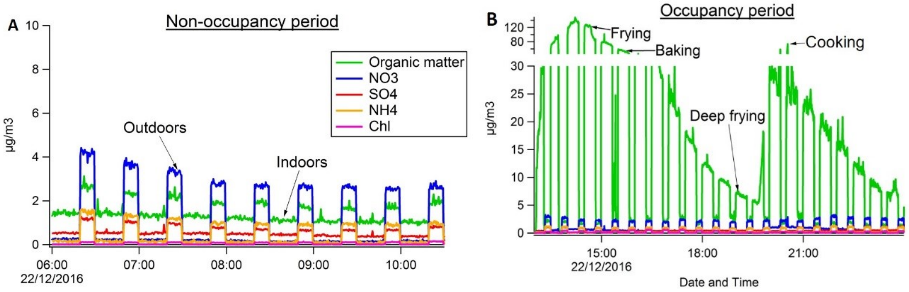


Figure 5. Particle mass concentrations of different chemical fractions during non-occupancy (A, 6:00–10:30), and occupancy periods (B, 14:30–24:00) inside and outside the apartment during one day

To illustrate the physicochemical transformations that occur during outdoor-to-indoor transport, a period without active indoor sources was chosen (Figure 5A). The indoor particle mass concentrations of all measured chemical species were lower compared to outdoors and all chemical species behaved differently upon infiltration indoors.

The semi-volatile ammonium nitrate (NH_4NO_3) and ammonium chloride (NH_4Cl) aerosols evaporated during outdoor-to-indoor transport (Figure 5A). It was previously found that ammonium nitrate (NH_4NO_3) and ammonium chloride (NH_4Cl) aerosols are highly susceptible to changes in temperature, relative humidity and the gas phase concentrations of ammonia, nitric acid, and hydrochloric acid (1, 50, 52-57, 110-113). During our measurements the average measured outdoor temperature T_{out} was 4.3°C (SD 1.8) and ranged from -8.8 to 9.7 °C. The average outdoor relative humidity RH_{out} was 86.5 % (±6) and ranged from 58 to 100 %. Indoors, average T_{in} was 24 °C (SD 0.4) and ranged from 20 to 26.1 °C. The average RH_{in} was 41.4 % (SD 3.7) and ranged from 27 to 50 %. Additionally, the losses of ammonium nitrate indoors could have resulted from sorption of the gas-phase to indoor surfaces (52). Sulfate and BC are non-volatile species, thus the infiltration of the particle mass of those species can be seen without phase change (Figure 5A).

Figure 5B illustrates the influence of the indoor sources on PM1 mass concentration. At 14:30, indoor concentrations of organics started to increase due to various cooking activities and the mass concentrations became higher indoors than outdoors. The indoor organic concentration reached maximum mass concentration of 140 µg m^{-3} during frying. Baking, deep-frying and cooking started after 15:00. Organic mass concentrations started to decrease either because the particle concentrations emitted from baking and deep-frying was lower compared to frying, or because particle mass started to disperse to other parts of the residence. In comparison to our measurements, Brunekreef et al. (114) showed similar maximum particle mass concentrations during cooking activities (200-300 µg m^{-3} in the particle size range of 10 - 300 nm).
The main contributor to the particle mass concentration was e-cigarette vaping, followed by cooking and outdoor contribution. The average particle mass concentration indoors was two times higher (15 µg m^{-3}) than outdoors throughout the measurements. The AER in apartment 5 was 0.52 which is in accordance with the Swedish good practice values (77). The kitchen fan was used on a regular basis when cooking. However, maximum particle mass concentrations reached high values during cooking (20 – 351 µg m^{-3}) and e-cigarette vaping (27-15700 µg m^{-3}).

The effect of energy renovation and occupants' activities on PM2.5 concentrations

This section describes the effects of energy renovation and occupants' activities on the UFP number and PM2.5 mass concentrations inside seven occupied Swedish apartments (apartment’s № in Table 1).

UFP number concentration. Figure 6A-C illustrates the differences in average UFP number concentrations of the individual apartments before, after renovation and at the follow-up during total monitoring time. The I/O ratios (also presented in Paper III, Table 1) of the UFP have decreased after renovation in apartments 8, 10, 12, 13, 15 in comparison to before renovation. However, the I/O ratios of the UFP in apartments 6, 13 and 15 at the follow-up were higher than before the renovation. Thus, the observed I/O ratios cannot be explained as being a result of the renovation and could rather have been influenced by the occupants' activities.

Figure 2 in Paper III shows frequency of indoor activities and the resulting UFP concentrations. It can be seen that the occupants performed on average two times more indoor activities during the follow-up compared to before and after renovation. Indoor activities, such as cooking/frying, candle burning and a combination of two or more than two activities emitted the highest UFP number concentrations during all the years measured. Most of indoor activities resulted in average UFP concentrations below 90 000 cm^{-3} and maximum UFP number concentrations were below 420 000 cm^{-3} during the entire measurement period (Paper III, Figure 2). Thus, considering the contribution from indoor activities to the UFP levels indoors, their frequency significantly affects the UFP levels indoors.

On average (Paper III, Figure S15), UFP concentrations decreased by 15% after renovation (5300 cm^{-3}, SD 4700 cm^{-3}) compared to before renovation (6200 cm^{-3}, SD 4000 cm^{-3}) in the studied apartments. The decrease in the UFP number concentrations after renovation can be explained by 1) increased AER (i.e., by an increase of bathroom exhaust airflows and more efficient kitchen hoods; 2) a decrease of indoor activities compared to before renovation.

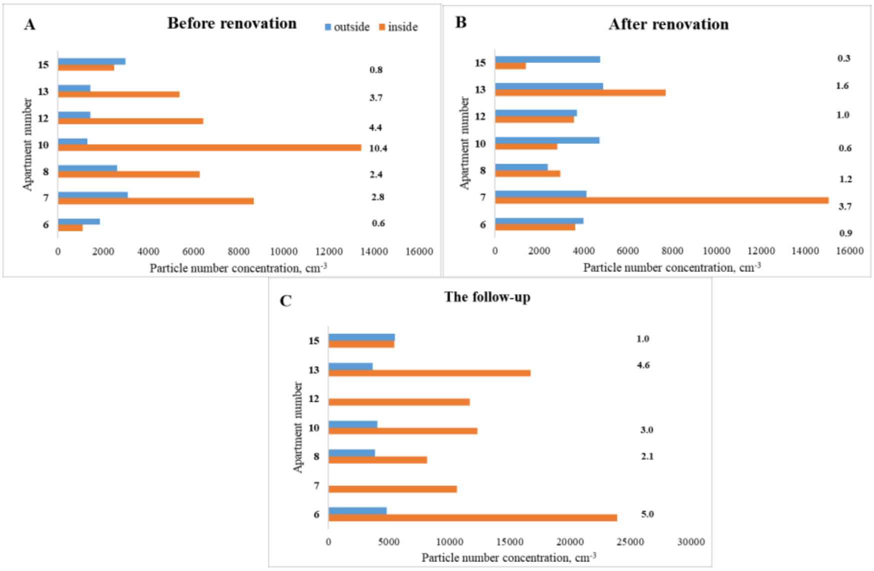


Figure 6. Differences in the average UFP number concentrations of the individual apartments before (A), after (B) and at the follow-up (C) and their respective outdoor concentrations (based on the measurements by NanoTracer). I/O ratios are specified on the right side of the charts.

During the follow-up, average UFP number concentration increased as the number of indoor activities doubled in comparison to before and after renovation. Cooking activities were the most typical indoor source, accounting for 49 % of all logged activities during the follow-up (Figure 3 in Paper III). Data from the magnetic sensor of the Activity Detection System (ADS) detected that the kitchen hood, which should help in more efficient removal of UFP, was used more frequently in the forced mode (80% of the time while cooking) at the follow-up compared to directly after renovation (67%). However, the kitchen hoods were not able to efficiently remove high concentrations of UFP generated during indoor activities. This indicate that kitchen exhaust flows need to be optimized to quickly and efficiently remove particles generated by cooking in order to reduce exposure to UFPs. This can be achieved by adopting more stringent building regulations for kitchen extraction hoods.

Particles of outdoor origin also contributed to indoor concentrations of UFPs to some extent. The average outdoor concentrations of UFPs were: 2100 cm^{-3}, SD 770 cm^{-3} before renovation; 4100 cm^{-3}, SD 900 cm^{-3} after renovation, and 4400 cm^{-3}, SD 1000 cm^{-3} at the follow-up. An illustration of the effect of UFP infiltration indoors, when indoor concentrations were lower than the respective outdoors concentrations during one-week of measurements is shown in Paper III in Figure S11B. To estimate the infiltrated particle fraction indoors, particle number concentrations were calculated during non-activity periods (i.e. when no one was in the residence, during nighttime (00:00-05:00), and during the time when the concentration was no longer influenced by preceding indoor activities (described in more detail in Paper III). The contribution of outdoor origin particles to the total average indoor number concentration was 23 % (1400 cm^{-3}) before renovation, 33 % (1700 cm^{-3}) after and 23 % (2900 cm^{-3}) at the follow up (Paper III, Figure S15).

Activity detection systems was used for detection of the occupants' activities during the measurements. The number of cooking activities reported in occupants' logbook records were compared to cooking activities detected by ADS and to the UFP data. The comparison showed underreporting or overreporting of the events by the occupants. This confirm the difficulty with identification of occupants' activities for particle source apportionment in indoor environments and the use of small, portable and accurate detection systems should be encouraged.

PM2.5 mass concentration. The I/O ratios of PM2.5 mass concentration in the apartments ranged from 0.8-2.6 before renovation and decreased to 0.1-0.6 at the follow-up (Paper III, Table 1). This corresponds to a 71% decrease of the average PM2.5 mass concentration indoors (2.5 µg m^{-3}, SD 1.3 µg m^{-3}) at the follow-up in compared to before renovation (8.6 µg m^{-3}, SD 5.8 µg m^{-3}) (Paper III, Figure S16).

The outdoor infiltration of PM2.5 mass concentration was lower after renovation and accounted for 30% (as the outdoor loadings were lower in that period) compared to before renovation (48%) (Paper III, Figure S16). However, at the follow-up indoor particle mass concentration was the lowest and infiltration from outdoors accounted to 7 % even though the outdoor mass concentration was the highest during three years of measurements.

The decrease in indoor PM2.5 mass concentrations can be explained by one, or by a combination, of the following: 1) lower amount of PM2.5 generated from indoor activities as the amount of activities decreased at the follow-up (N=7 amount of activities identified as increasing PM2.5 levels out of a total of 40 activities), (Paper

III, Figure S13); 2) decreased infiltration of outdoor particles as a result of the renovation, that is, the installation of the small ventilation vents (instead of the window-like openings in the living rooms and bedrooms that reduced the cross-section area for outdoor supply air flow and increased the area where particles could deposit via impaction), and tightening of the building envelope (by the addition of extra insulation in the building wall, the sealing of the windows, and the replacement of curtain walls and windows on the south façade, explained in more detail in Paper III).

The differences in PM2.5 mass concentrations and black carbon between individual apartments are shown in Paper III on Figure 4. In some apartments, PM2.5 mass concentrations were higher or lower compared to outdoors during all measurement rounds. Higher indoor PM2.5 mass concentrations can be explained by the presence of indoor sources in addition to infiltrated particles of outdoor origin. Lower indoor PM2.5 mass concentrations indicated that indoor concentrations were mostly influenced by the outdoor infiltration. The sources of black carbon were outdoor infiltration, candles and incense burning, frying and smoking (Paper III, Figure 4).

Characterization of particle emissions from burning of candles

Time series and emission factors. The particle emissions of five different pillar candle types (Paper IV, Table 1) under stressed burning conditions were dominated by soot (black carbon, eBC), UFP number, and PM2.5 mass concentrations. Each candle had different profiles in terms of number and mass emissions. The UFP number concentration (measured between 3–80 nm) in the chamber varied between 2×10^6 –5×10^6 cm^{-3} over the entire time of the experiments. A peak in UFP number concentration appeared directly after the candles were lit, except from candle 3 that showed more stable emissions.

The different candles had different eBC emissions. Candles 3 and 5 showed a gradual increase of eBC over time. The mass concentration of eBC from candle 5 was however, thirteen times higher compared to that from candle 3. Candle 4 showed a peak emission of eBC at the beginning of the burning that declined after 1–1.5 h. Emissions from candles 1 and 2 (Paper IV, Figure S2) were relatively similar during the initial half hour, but after that, the eBC mass concentrations from both candles started deviating from each other and fluctuating.

Table 7. Emission factors of different indoor sources based on UFP number and PM2.5 mass concentrations, BC online measurements.

	PNC (2.4-100 nm)		PM$_{2.5}$[6]		eBC	
	#/h	SD	µg/h	SD	µg/h	SD
Candle 1[1]	9.3E+13	$\pm0.2\times10^{13}$	283	±53	408	±50.5
Candle 2[1]	9.0E+13	$\pm0.4\times10^{13}$	510	±61	634	±0.5
Candle 3[1]	6.3E+13	$\pm0.3\times10^{13}$	393	±57	329	±89.5
Candle 4[1]	8.5E+13	$\pm0.4\times10^{13}$	330[4,5]		160	±68.5
Candle 5[1]	1.3E+13	$\pm0.1\times10^{13}$	3038	±1691	4346	±874
Candle 1 (steady)[2]	8.0E+13		<DL	<DL	2.6	
Candle burning in homes[3]	3.4E+12	$\pm5.7\times10^{12}$				

[1] Emission factors (EFs) of PNC for candles 1-5 under stressed burn based on 3 h average concentrations due to use of the catalytic stripper in the last 30 min of the experiment.

[2] EFs for the steady candle experiment in the laboratory chamber, based on a single measurement

[3] Average of 15 candle burning events from the measurements in 15 homes (size range 10-300 nm)

[4] Possibly underestimated value, because the peak emissions occurring within the first half hour are not included (sampling 0.5-3h)

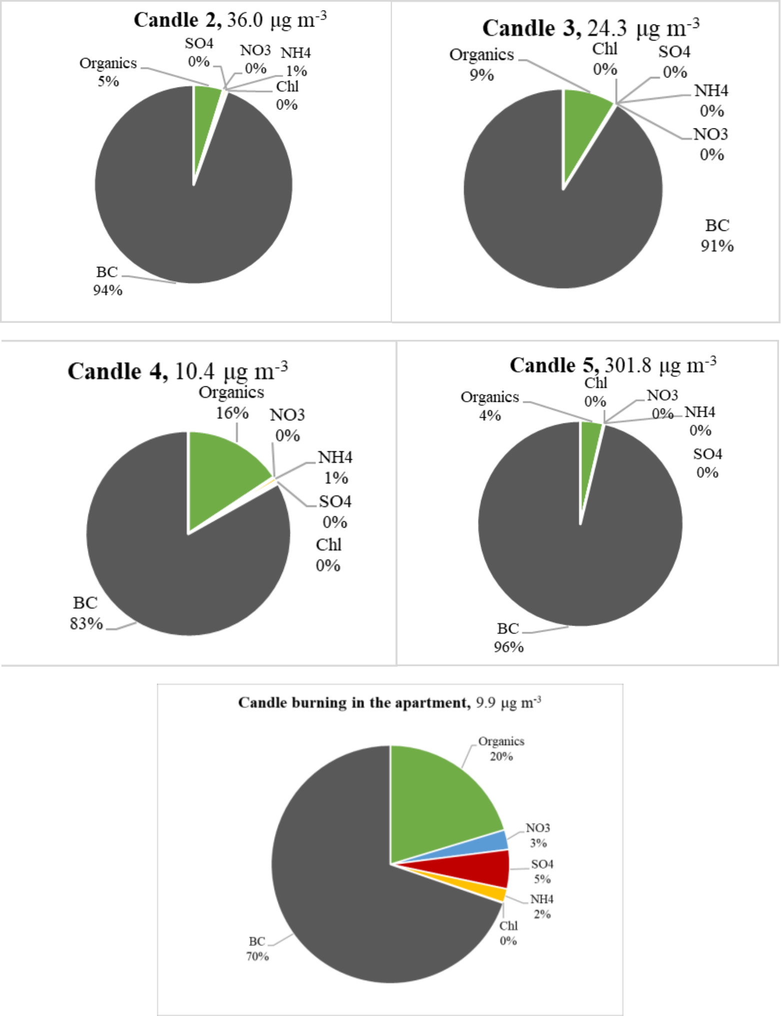
[5] Value based on a single measurement

[6] Based on the gravimetric analysis

Emission factors (EFs) of UFP number, PM$_{2.5}$ and eBC mass were calculated for each of the candle types over 3 h (Table 7). The EFs of UFPs (2.4–100 nm) were similar for all candles (Table 7), however, the emissions of candle 5 were the lowest (1.3×10^{13} particles per h). EF of UFPs (10–300 nm) during candle burning in the occupied homes were slightly lower compared to the laboratory measurements. The difference in the EF can be explained by the different type of candles used by occupants, burning conditions measured size and deposition on the indoor surfaces. In real indoor environments, there are more surface area available for particle deposition, thus deposition is more enhanced (63).

Candle 5 had the highest EFs eBC and PM2.5 among all candles but the lowest EF of UFP that was most probably due to coagulation of ultrafine particles with soot particles.

Candle burning was found to emit UFP, black carbon, PM2.5 and inorganic compounds, both, in the laboratory experiments as well as in 15 homes. In the laboratory experiments candles were burned under stressed and steady burning conditions. In a real indoor environment, it is more likely for a candle to burn under stressed conditions when the flame is affected by air movements (for instance, from an open window). In some cases, a candle can burn steady if the flame is undisturbed or if specific candle holders, that shield the flame from air movements, are used.


Candle 2, 36.0 µg m^{-3}

Candle 3, 24.3 µg m^{-3}

Candle 4, 10.4 µg m^{-3}

Candle 5, 301.8 µg m^{-3}

Candle burning in the apartment, 9.9 µg m^{-3}

Figure 7. Mass concentrations of organics, ammonia, sulfate, nitrate, chloride and black carbon (BC) in PM1 during candle burning in the occupied apartment (3.25 h average) and the laboratory experiments (2.5 h-average)

Figure 7 illustrates the chemical composition of PM1 during candle burning under stressed burn in the laboratory conditions and in home 5. It can be seen that PM1 chemical composition was dominated by eBC, followed by a small contribution of organics (4-16% of determined PM1 mass in laboratory study). The total PM1 mass concentrations varied between different candles in the laboratory and in home 5, however, the measurements were done under different conditions and space volumes.

The low mass of the inorganic compounds (in laboratory study candle 2, 3, 4 and 5 denoted as 0 - 1%) could be due to that inorganics are found in the ultrafine particle mode, i.e. with sizes below 100 nm (the mass-weighted mode diameter was 30–50 nm) thus not detected by AMS. This is because, the AMS has a limitation in measuring these particle sizes and of a cut-off diameter of is around 50 nm. In the occupied home, the inorganic concentrations detected could be a result of particle infiltration from outdoors, i.e., from background concentrations indoors. Higher fraction of organics in home 5 were also observed that could be a result of presence of residual concentration from previous indoor activities or infiltrated from outdoors.

Particle number and mass size distributions. Average particle number and mass size distributions (size range 2.4–914 nm, during 3 h) and replicate experiments of candles 1-5 during stressed burn during experiments in the chamber are illustrated in Paper IV (Figures 2, S6). The particle number size distributions of candles 1–4 were unimodal with a majority of the emitted particles in the UFP size range (average mode diameters between 5 and 8 nm) during stressed burn. Candle 5 showed a bimodal size distribution with one peak at 6 nm and one at ~200 nm. The second mode was formed by larger soot particles. The 'soot mode' during stressed burn was previously reported by Pagels et al. (12) (modes at~20–30 and ~300 nm) and by Wallace et al. (13) for citronella candle (modes at ~50 and ~200 nm). The chemical composition of the UFPs was investigated with help of a catalytic stripper (CS) in combination with nano-SMPS and off-line chemical analysis. The major components were found to be water-soluble inorganics such as phosphates, sulfates and nitrates.

Average particle mass size distributions were bimodal for candles 1–4 with a UFP mode between 27-44 nm and a larger soot mode between 550 - 900 nm (Paper IV, Figure 2, Figure S6). Bimodal (main mode at 290 nm and minor at 50 nm) mass size distribution was also observed by Stabile et al. (115) during burning of paraffin wax candles. Candle 5 in our study showed a unimodal mass size distribution with a peak at 570 nm.

In conclusion, candles emit high numbers of particles in the UFP size range both during stressed and steady burn in our laboratory experiments. The particle chemical composition of all candles was dominated by eBC, with a small contribution of inorganic and organic components. The emissions of eBC, PM2.5 and particle-phase PAHs were influenced by the wax and wick composition and to lower degree ultrafine particles, inorganic and organic carbon fraction of PM. Candles are widely used in homes, especially in Scandinavian countries. Emissions of high mass concentration of eBC, PM2.5 and UFP number concentrations are of concern from a health perspective and may pose potential risks in relation to the human health.

Conclusions

The differences in physical, chemical and toxicological characteristics of fine particles have been assessed inside and outside occupied homes. As the measurements were performed in homes that were occupied, assessed particle concentrations reflect the contribution of both indoor and outdoor particle sources. Additionally, a chamber study was performed to assess emissions from candles, as these were identified as strong contributors to particle levels indoors. Among apartments that underwent energy renovations, particle concentrations were assessed before and after renovations.

Average UFP number concentrations were three times higher indoors (8600 cm^{-3}, SD 9300 cm^{-3}) compared to outdoors (2800 cm^{-3}, SD 1200 cm^{-3}) during the measurements in 15 homes. This can be explained by the presence of the indoor sources such as cooking (frying, boiling, baking), candle and incense burning, and vaping an e-cigarette. Particle emissions depended on the type and duration of the particle source. PM2.5 mass concentrations were similar indoors (7.5 $\mu g\ m^{-3}$, SD 6.0 $\mu g\ m^{-3}$) and outdoors (7.3 $\mu g\ m^{-3}$, SD 5.0 $\mu g\ m^{-3}$). However, a majority of the PM2.5 mass indoors infiltrated from outdoors (64 %) in the 15 homes. Some indoor sources also contributed to PM2.5 mass, however, indoor activities contributed less to PM2.5 mass than to UFPs in studied homes.

It was found that the average metal concentrations in PM2.5 was higher in 15 homes compared to outdoor concentrations. Metals originated from both outdoors and indoors. The dominating metals indoors were Fe, Cr, Al, Zn. and Mg. These particular metals were, apart from outdoor contribution, most likely emitted by sources such as cooking, candle and incense burning. The average mass concentrations of inorganics showed to be lower indoors compared to outdoors. However, inorganic species such as P, Ca, K, Na, Cl that were higher in some homes than others, partly originated from outdoors but also could have been emitted during candle burning or cooking. The average levels of PAHs were low both indoors and outdoors and a slightly higher average PAH concentration was found outdoors. The average endotoxin concentration was higher indoors in comparison to outdoors, however, the found concentrations did not exceed the occupational exposure limit values.

Compared to outdoor particles, higher toxicity was found in indoor particles assessed by in-vivo toxicological exposure in mice. This is most probably associated with a higher amount of metals, PAHs and endotoxins in the extracted indoor particles in comparison to outdoor particles. It should be pointed out that, in the particles used for toxicological studies, determined PAHs concentrations indoors were higher than outdoors, which was opposite to was found on individual filters. This was most probably due to lower temperature at the indoor collection point for toxicological studies (i.e. outside the sampling case) in comparison to all other samples. This lower temperature most probably reduced evaporation and hence preserved PAHs in particle phase.

A detailed online chemical characterization with HR-ToF-AMS of PM1 mass in home 5 showed that emissions were dominated by organic matter emitted during cooking activities and e-cigarette vaping. Organic PM1 mass was lower outdoors compared to indoors. Emissions of black carbon (eBC) mass were observed during cooking (e.g., frying) and candle burning in that home.

Chamber experiments assessing candle burning under stressed conditions also showed that emissions were dominated by eBC. There were variations in emission factors of PM2.5, eBC and particle-phase PAHs between different candles types and it was proved that these were influenced by differences in the wax and wick composition.

After the energy renovation, the average indoor PM2.5 mass concentration decreased from 8.6 µg m^{-3} (SD 5.8) before renovation to 2.5 µg m^{-3} (SD 1.3) compared to the follow up measurements. This can be due to two reasons: an observed decrease of PM2.5-generating indoor activities, and a decrease in infiltration of outdoor particles due to the renovation measures. However, such a strong decrease in outdoor infiltration was not observed after the renovation in comparison to before renovation, hence the decrease during follow up should be treated with caution. Indoor UFP number concentrations depended mainly on frequency and type of occupants' activities (e.g., cooking, candle burning). UFP number concentrations indoors increased during the follow up compared to before renovation, as the amount of activities doubled. Newly installed kitchen extraction hoods were used more frequently in forced mode while cooking during the follow-up measurements compared to during the after renovation measurements. This did not, however, efficiently remove high particle concentrations generated indoors. Thus, in order to reduce personal exposure to indoor UFP, optimization of kitchen exhaust flows should be considered.

For future studies for accurate particle source apportionment, there is a need of development of small, portable sensors that would enable monitoring pollutants level (e.g., particle concentration), indoor environment quality parameters (e.g. temperature, relative humidity) and occupants activities (e.g. use of stove and oven, window opening).

The knowledge obtained in this thesis can be used for developing strategies to minimize particle exposure indoors by combination of effective particle removal indoors and reducing outdoor infiltration. The current data can be used in mapping of real-life indoor concentrations and for modeling of exposure assessment.

Outlook

The results from toxicological studies in mice in the present thesis showed higher toxicity of indoor particles associated with higher amount of metals, PAHs and endotoxins in extracted particles than outdoor particles. The metals originated from both outdoor and indoor sources and the dominant metal element indoors was iron. Previous studies have associated iron with generation of reactive oxygen species (ROS) that can cause damage to DNA and induce deleterious health effects (93-95). Studying ROS properties of indoor particles could help in guiding towards understanding toxicological effect.

Found results underline the importance of continuing studying health effects of particles in indoor environments. An example of possible future research is conducting toxicological studies based on the collected particles in real indoor environments (e.g. occupied homes) and investigating chemical composition of indoor particles. Understanding health effects from the particles found indoor is of particular importance as we spend majority of our time indoors. Like in study conducted in this thesis but on larger scale, i.e. covering larger variability of homes in different regions.

The above-mentioned studies need to be supported by detailed monitoring of occupant activities in order to perform particle source apportionment in indoor environments. There is a need of using small and portable sensors that would provide data about different parameters (e.g., particle concentration, relative humidity, temperature, movement). This would enable monitoring those parameters in many indoor environments.

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