



**KAUNAS UNIVERSITY OF TECHNOLOGY
FACULTY OF CHEMICAL TECHNOLOGY**

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**DIFFERENCES IN PARTICLE PHYSICO-CHEMICAL
PROPERTIES INSIDE AND OUTSIDE AN OCCUPIED
RESIDENCE**

Master's Degree Final Project

Supervisors

Assoc. prof. dr. Aneta Wierzbicka

Assoc. prof. dr. Dainius Martuzevicius

KAUNAS, 2017

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Environmental Engineering (code 621H17001)

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"Differences in particle physico-chemical properties inside and outside an occupied residence"

DECLARATION OF ACADEMIC INTEGRITY

2017 June 9
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The present Master thesis has been a part of a research project «Airborne particles in our homes: cocktail effects, chemical composition, physical characteristics and toxicity» (Formas funded project Dn 942-2015-1029) and was done in collaboration between Lund University and Kaunas University of Technology within Erasmus Traineeship program.

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SUMMARY

A number of health effects have been linked to exposure to airborne particles, including respiratory symptoms, cardiovascular diseases, and increased mortality. These are further complicated by the fact that people are spending more time indoors, in particular, almost 69 % we are spending in homes (EPA), where particles of indoor and outdoor origin are found. The aim of the present Master thesis was to assess the differences in physico-chemical particle characteristics inside and outside of the occupied residence and to identify the origin of major contributors to the observed levels indoors.

Simultaneous, indoor and outdoor measurements were performed in naturally ventilated four-bedroom apartment in Malmö, Sweden. Scanning Mobility Particle Sizer (SMPS) and Time-of-Flight Aerosol Mass Spectrometer (AMS) were used to measure particle number and mass concentrations, and size-resolved distributions inside and outside of the apartment. Automatically switching valve alternated between indoor and outdoor lines with the time resolution of 20 and 10 minutes, respectively. Both sampling lines were mounted at the ground floor level and led to the setup where the aerosol was dried and measured by both instruments. Indoor sampling line was heated and insulated to keep the same temperature of the indoor particles and additional carrier flow was used to lower the residence time.

Results have showed that indoor particle loadings, in terms of particle number and mass concentrations, substantially exceeded the outdoor values due to the contribution of indoor

emission sources during occupancy period, low air exchange rate during winter season in naturally ventilated apartment, and variations of outdoor particle concentration. The main contributors to indoor loadings were identified to be various types of cooking (frying, deep frying, baking, toasting, breakfast preparation), candle burning, ironing of the clothes. Number particle size distribution of the individual indoor sources showed to be distributed in ultra-fine particle size range (UFP, <100 nm) which made up 84% of total particle number concentration (PNC) within occupancy period, i.e. when at least one person is present indoors. This is important in relation to human health since UFP are known to have higher deposition rates in the lower respiratory tract, compared with larger particles. The period without indoor activities (non-occupancy period) showed the influence of the outdoor particles, but this period was affected by the concentration generated previously from cooking activities and particle loadings were overestimated during this time. Outdoor loadings were influenced by traffic and some local sources, such as emissions from fireplaces (used by neighbours) and from adjacent fast food restaurants.

Indoor to outdoor (I/O) ratios on the basis of particle mass of chemical species have accounted: for organic matter 3.4 due to the emissions of primary PM from indoor activities, and possibly, formation of secondary organic species took place; for ammonium 0.13 and for nitrate 0.22 as semi-volatile ammonium nitrate is sensitive to the temperature and relative humidity differences, reflecting phase change upon outdoor-to-indoor transport; for sulphate 0.51 and chloride 0.25, which have showed a result of outdoor penetration assuming the absence of indoor sulphate sources. However, emissions of nitrate and chloride may have resulted from episodic sources, such as candle burning, alkali nitrate salts and zinc chloride which were possibly added as flame retardants to the candle wick. The emitted nitrate could be another precursor to the formation of secondary organic species.

The present analysis of collected data from the state-of-the-art instruments reflects the real-time indoor concentrations of specific components which can be the indicators of indoor air quality. Considering substantial amount of found submicrometer-size range particulate matter of different chemical composition indoors, originated from indoor and outdoor sources, creates potential risk in relation to the human health. Further detailed data analysis will provide more information about relative contribution of different components and origin of PM and air quality management strategies will be found in order to help in achieving suitable indoor air quality.

Omelekhina, Yuliya. AEROZOLIO DALELIŲ FIZIKINIŲ-CHEMINIŲ SAVYBIŲ SKIRTUMAI APGYVENDINTŲ PATALPŲ VISUJE IR IŠORĖJE: Aplinkos inžinerijos magistro darbas / vadovai doc. Aneta Wierzbicka, Inžinerijos fakultetas, Dizaino mokslų departamentas, Lundo universitetas; doc. Dainius Martuzevičius, Aplinkosaugos technologijos katedra, Cheminės technologijos fakultetas, Kauno technologijos universitetas.

Magistro darbas parengtas mokslinių tyrimų projekto „Aerolio dalelės namuose: mišinių poveikis, cheminė sudėtis, fizinės savybės ir toksiškumas“ (Projekto nr. Dn 942-2015-1029, Švedijos mokslo taryba Formas) rėmuose, bendradarbiaujant Lundo universitetui ir Kauno technologijos universitetui Erasmus Stažuočių programos metu.

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Raktiniai žodžiai: Patalpų oro kokybė, aerolio dalelės, dalelių fizinė ir cheminė sudėtis, Skanuojantis dalelių mobilumo matuoklis, Skriejimo laiko aerolio masių spektrometras, patalpų ir išorės teršalų santykis.

Lundas, Kaunas, 2017.

SANTRAUKA

Ekspozicija ore esančioms dalelėmis yra siejama su neigiamu poveikiu sveikatai, įskaitant kvėpavimo takų, širdies ir kraujagyslių ligomis bei padidėjusiu mirtingumu. Papildomų problemų vertinant ekspoziciją sukelia faktas, kad žmonės didžiąją laiko dalį (beveik 69%, EPA duomenys) praleidžia patalpose, kuriose aptinkama tiek vidaus, tiek išorės kilmės dalelių. Šio magistro darbo tikslas buvo ištirti fiziko-cheminių dalelių savybių skirtumus apgyvendintų patalpų viduje ir išorėje bei identifikuoti pagrindinius dalelių šaltinius patalpose.

Vidaus ir patalpų oro užterštumo matavimai buvo lygiagrečiai atliekami natūraliai vėdinamame keturių miegamųjų bute Malmėje, Švedijoje. Skanuojantis dalelių mobilumo analizatorius ir skrydžio laiko aerolio masių spektrometras buvo naudojami matuoti dalelių skaitinei ir masės koncentracijai bei dispersinei sudėčiai buto viduje ir išorėje. Automatinis vožtuvas, veikiantis su 10 ar 20 minučių skiriamąja geba leido vienu metu imti išorės ir patalpų oro ėminius. Abi ėminių ėmimo linijos buvo sumontuotos pažemiui ir vedė į rūšį, kur aerolis buvo išdžiovintas ir matuojamas abiem instrumentais. Patalpų oro linija buvo šildoma ir apšiltinta, siekiant išsaugoti daleles toje pačioje temperatūroje. Papildomas nešančiojo oro srautas buvo naudojamas sumažinti pernešimo iki instrumentų laiką.

Matavimų rezultatai rodo, kad patalpų oro apkrova dalelėmis vertinant pagal skaitinę ir masės koncentraciją, reikšmingai viršijo išorės vertes dėl patalpų šaltinių įtakos patalpoje esant

gyventojams, taip pat dėl mažesnės oro apykaitos šaltuoju metų sezonu bei išorės oro dalelių koncentracijų kitimo. Pagrindiniai nustatyti teršalų šaltiniai patalpų viduje buvo įvairios maisto gamybos operacijos (gruzdinimas, gilusis gruzdinimas, kepimas, duonos apkepinimas, pusryčių ruošimas), žvakių deginimas, drabužių lyginimas. Šių šaltinių dalelės pagrinde buvo sudarytos iš organinių medžiagų. Atskirų šaltinių dalelių skaitinė koncentracija ir dispersinė sudėtis daugiausia buvo pasiskirsčiusi ultra-smulkiųjų dalelių intervale (<100 nm), kuris sudarė iki 84 % visų dalelių skaitinės koncentracijos, kai patalpos buvo naudojamos. Tai yra svarbus indikatorius, kadangi ultrasmulkiosios dalelės yra siejamos su poveikiu sveikatai dėl didesnio nusėdinimo apatiniuose kvėpavimo takuose, palyginus su didesnėmis dalelėmis. Periodais, kai patalpos nebuvo naudojamos, išryškėjo išorės dalelių įtaka, tačiau taip pat buvo pastebima prieš tai vykusių patalpų taršos epizodų (maisto gamybos), kas sąlygojo apkrovos pervertinimą. Išorės apkrovos buvo įtakojamos gatvių eismo ir kelių vietinių šaltinių, kaip židinių ir netoliese esančių restoranų emisijos.

Dalelių sudedamųjų dalių vidaus ir patalpų koncentracijų santykis amonio jonų atveju buvo 0,13, nitrato 0,22 dėl pusiau lakaus amonio nitrato priklausomybės nuo temperatūros ir drėgmės pokyčių transportuojant aerozolį ir į patalpas. Sulfato jonų atveju santykis siekė 0,51, ir parodė išorės dalelių prasiskverbimą, priimant, kad patalpose nebuvo sulfatų šaltinio. Chloridų atveju santykis sudarė 0,25. Antra vertus, chlorido ir nitrato jonai galėjo būti išmetami patalpų šaltinių, tokių kaip žvakių deginimas, įtakojantis šarminių metalų nitratų druskų bei cinko chlorido emisiją kaip degimą sunkinančių medžiagų žvakių dagtyje. Nitrato jonas taip pat galėjo pasitarnauti kaip prekursorius antrinėms aerolio dalelėms susidaryti.

Naujausių aerolio savybių matavimų instrumentų pagalba surinkti duomenys davė žinių apie realaus laiko kai kurių aerolio komponentų koncentracijų kitimus, kurie yra svarbūs patalpų oro kokybės indikatoriai. Reikšmingas įvairios cheminės sudėties submikrometrinių dalelių, atsirandančių tiek iš patalpų, tiek iš vidaus šaltinių, kiekis patalpų viduje leidžia daryti išvadą dėl jų potencialaus pavojaus žmonių sveikatai. Duomenys turi būti toliau analizuojami siekiant nustatyti įvairių komponentų įtaką ir dalelių kilmę, bei pateikiant rekomendacijas apie šių duomenų naudojimą oro kokybės valdymui pastatuose.

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INTRODUCTION

Throughout their entire lives, each and every person are subjected to particulate matter present in indoor air [52]. The airborne particles can be taken up by the occupants: they can either be inhaled when building up in air or become resuspended, or ingested if they are carried to the human mouth or to food. Upon entering the human body airborne particles create potential health hazards. Indoors, particles often come from indoor sources, infiltrate from outdoor air or result in new particles formation during chemical reactions between precursors of indoor and outdoor origin. A number of health effects have been identified as being linked to exposure to airborne particles, including respiratory symptoms, cardiovascular diseases, and increased mortality. These potentially adverse health effects are further complicated by the fact that people are spending more time than ever indoors, up to 90 percent according to estimates by the U.S. Environmental Protection Agency (EPA). It is easy to understand why there is a growing concern about the quality of the air we breathe. It was assessed that 10–30% of the total burden of disease from particulate matter exposure was due to indoor-generated particles, signifying that indoor environments are likely to be a dominant environmental factor affecting human health. Health, environmental, and other effects caused by particles are the reasons for monitoring and controlling their presence in the indoor environment. Thus, an understanding has to be provided about particles origins, the contribution from individual sources, their characteristics, behaviour, and the effects they may cause.

The main aim of the present Master thesis was to assess differences in physico-chemical characteristics of the airborne particles between outdoor and indoor environments.

Specifically:

- to design the setup and perform simultaneous, detailed time and size resolved particle measurements inside and outside a residence in Malmo city, Sweden;
- determine differences in physico-chemical particle characteristics between indoor and outdoor by using number and mass concentration, number size distribution, indoor-to-outdoor ratio
- Identify origin of major contributors to observed levels indoors.

1. BACKGROUND AND THEORY

1.1 AEROSOL DEFINITION

Aerosol is known as an assembly of liquid or solid particles suspended in a gaseous medium long enough to be observed or measured [49].

1.2 PARTICLE SIZE AND SHAPE

Particle size largely determines the particle motion in gas suspension, as well as describes particle behaviour in the respiratory tract. The size of aerosol particles spans in diameter from 0.001 μm to 100 μm (10^{-9} to 10^{-4} m).

According to the size spectrum particles can be divided into three modes: ultrafine ($< 0.1 \mu\text{m}$), fine (0.1-2.5 μm), and coarse ($> 2.5 \mu\text{m}$). Depending of the size of the particle different physical laws apply. For instance, Brownian diffusion is the main deposition mechanism for particles less than 0.1 μm . While particles larger than 1 μm are effectively removed by gravitational settling, and particles with diameters $< 0.1 \mu\text{m}$ are able to diffuse through the air, and thus end up on surfaces, particles with diameters in the range of 0.1 μm to 1 μm are not effectively removed by either mechanism.

Inhaled particles of different physico-chemical nature may deposit in various region of the respiratory system. During inhalation, the incoming air undergoes a series of changes in direction as it flows from the nose to the mouth down through the alveolar region. The Brownian motion of submicrometer-sized particles leads to increased likelihood that they will deposit in the airway walls where distances are short and residence times comparatively long [49].

Liquid particles are nearly always spherical while solid ones may have complex shapes. In studying particle properties, the assumption that all particles are spherical is made. An equivalent aerodynamic diameter is frequently used, which is the diameter of a standard density sphere (1000 kg/m^3) that has the same aerodynamic properties as has an irregular particle. When a particle is determined by a given technique, the measurement corresponds to a specific measured physical property. In an electric field, a particle of known charge moves along a predictable trajectory. Thus, the motion of a particle is of concern, the mobility equivalent diameter is used, which is the diameter of a sphere with the same electrical mobility as particle in question [48, 49]. The time-of-flight based particle measuring technique operates by creating particle velocities in a response to the pressure difference and provide means of obtaining the aerodynamic diameter by measuring particle time of flight [22].

1.3 PARTICLE CONCENTRATIONS

Particle number concentration (PNC) is the number of particles per unit volume of aerosol (cm^{-3}) which is commonly expressed as number cm^{-3} . Another measure of concentration is *mass concentration (PMC)* of particulate matter in a unit volume of aerosol (m^{-3}). The amount of particle mass is expressed in g m^{-3} , mg m^{-3} , $\mu\text{g m}^{-3}$.

1.4 PARTICLE SIZE DISTRIBUTION

Physico-chemical properties of particles are size dependent. The result of size analysis might be a great number of particle sizes and a very convenient way of handling the variability in size is through the size distributions. The size distribution can be represented by plotting particle mass or number versus size. Size distribution is obtained by dividing total mass or number particle concentration in the range by the difference between the log of the upper boundary and the log of the lower bin boundary in each channel (normalizing for bin width) [49].

$$\frac{dN}{d\log D_p} = \frac{dN}{\log D_{p,u} - \log D_{p,l}} \quad (1.1)$$

where:

dN - particle concentration

D_p - midpoint particle diameter

$D_{p,u}$ - upper channel diameter

$D_{p,l}$ - lower channel diameter

2. BEHAVIOUR, TRANSPORT, AND FATE OF PARTICLE IN INDOOR ENVIRONMENT

Airborne particulate matter is a diverse pollutant class which in excessive amounts in indoor air may contribute to an adverse health effects. Since people are spending most of their life indoors (in many regions more than 90%), and it is easy to understand that the important environment in relation to our health is the indoor environment [55].

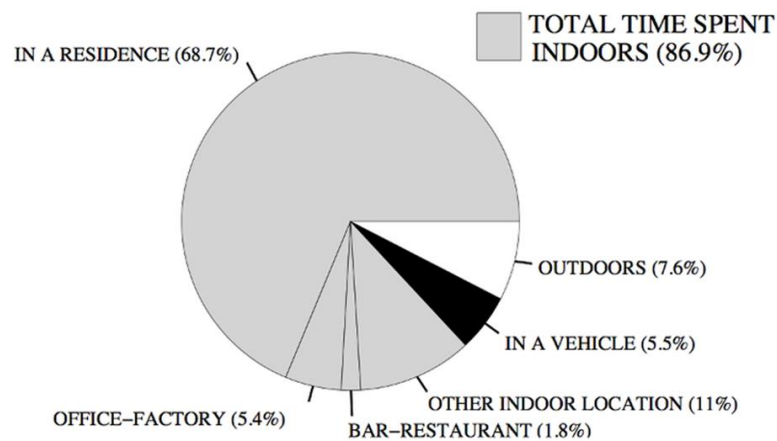


Fig. 1.1. Time spend in different locations (National Human Activity Pattern Survey (NHAPS), 2001)

The significance of PM to indoor air quality is strongly related to airborne particle concentration, size distribution, and chemical composition, which in turn, depend on sources, removal mechanisms, and transformation processes (Figure 1.2).

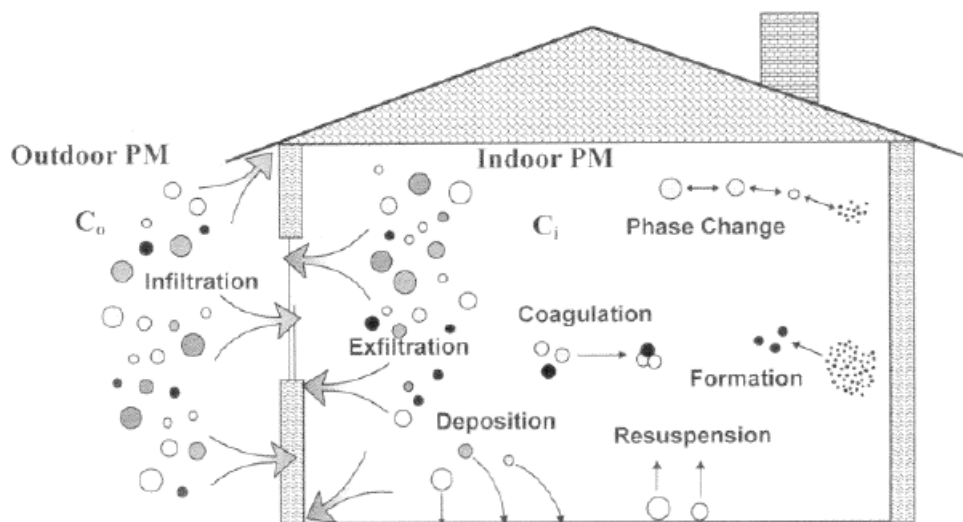


Fig. 1.2. Behaviour, removal mechanisms and transformation processes in indoor environment [57]

Particle concentrations indoors could be affected by the following sources and processes:

- indoor emissions sources (related to human activity);

- chemical reactions involving vapours and gases leading to particle generation;
- penetration of outdoor particles indoors through open doors and windows, as well as through the building envelope;
- resuspension of particles deposited on surfaces;
- removal of particles from the indoor environment by ventilation;
- deposition of particles originating from both indoor and outdoor sources on indoor surfaces [53].
- other factors: building characteristics, geographical location, hobbies, type of interiors [58].

2.1 INDOOR SOURCES OF PARTICLES

2.1.1 Emissions from cooking activities

Cooking activities, are the concurrent part of everyday routine. When food is cooked at high temperatures (up to 300°C) degradation of carbohydrates, fats and denaturation of proteins occurs resulting formation of *ultrafine (UFP)* and *fine particulate matter, organic gaseous emissions, secondary organic aerosols*, and *steam* from the water [3; 27; 10; 9; 8; 11; 6; 12; 1; 7; 13; 39; 24; 14; 5; 2; 15; 60; 64]. Coarse particles were also found during sauteing [61].

During food preparation, *particulate matter* is formed by the cooling of hot vapour created by evaporation of organic constituents from cooking products (i.e. cooking oils) upon heating above its boiling point [39]. However, particle concentrations which are being emitted from different types of cooking activities may vary. For instance, during frying, particle number concentrations (PNC) at the main peak, particles cm^{-3} , were: for boiling 56 000, frying 14 000, baking 230000, toast making 160 000 [2].

[9] studied formation of fine and ultrafine PM from cooking, such as, frying (frying eggs in butter, stir-frying in a wok), deep-frying (frying flour tortillas in peanut oil), baking, toasting, sauteing (of Swiss chardin), grilling (veggie burgers) on a gas stove in an occupied townhouse. Particle size distribution was bimodal with a peak at about 10 nm with particle number concentration of 1500 particles cm^{-3} , and a second peak at about 60 nm, with 3300 particles cm^{-3} . Total mass produced was 50 $\mu\text{g m}^{-3}$ dominated by particle size range of 100 - 300 nm (comprise 30 % of mass) over a typical cooking time of about 5-15 min. During stir-frying on a gas burner have produced a peak of PM at ~35 nm, whereas deep-frying on the gas burner followed by baking produced a peak about twice as high at ~64 nm. It was also noted that the gas flame itself produces ultrafine particles. When burners were on with no pot or food being cooked, large numbers of

ultrafine particles were produced, with smallest peak of 9.8 nm. The electric toaster oven could produce ultrafine particles peaking at about 30 nm when no bread was inserted.

[63] describe PNCs at the peak values to be 590,000 particles cm^{-3} (<100 nm) during frying of bacon on a gas stove.

[39] found large amounts of fine particulate matter (PM_{2.5}) was produced from heating cooking oils. Emission rate was shown to be 54 mg min^{-1} for peanut, canola, corn and olive oils heated at 197 °C.

Cooking activities may also lead to the formation of *secondary aerosols*. The emitted from cooking volatile organic compounds, such as, formaldehyde, acetaldehyde, acrylamide, acrolein [27] in presence of ozone, hydroxyl or nitrate radicals may cause reactions resulting in formation secondary organic aerosols.

Found indoors ozone mostly originate from outdoor environment [28] depending on type of ventilation system (mechanical or natural) used. However, there are some direct indoor emissions of ozone generated from printers and photocopiers [30]. Some studies suggest that naturally ventilated buildings tend to have higher ozone concentrations comparing to those with air conditioning systems. HVAC (heating, ventilation and air condition) systems may reduce concentrations indoors with filters by 22-95% of the total removal. Moreover, a building with a lower air exchange rate (AER) has lower ozone concentrations. It could also be expected to have larger ozone concentration in summer months, compared to winter period, when windows are open more frequently [42]. In Nordic countries, the ozone concentrations are lower compared to the other parts of the Europe [51]. Hydroxyl radicals and nitrate radicals are other oxidants that may be present indoors at sufficient concentrations to produce secondary pollutants. Hydroxyl radicals are produced during reactions of ozone with a various VOCs [28], and indoor nitrate radicals result from the reaction between ozone and nitrogen dioxide.

Contribution of cooking emissions to formation of secondary organic aerosol (SOA) from gas-phase emissions of heated vegetable oils (i.e. corn, canola, sunflower, peanut and olive oils) was studied [40] with high-resolution time-of-flight aerosol mass spectrometer (HR-TOF-AMS). Ozone was generated to the chamber at 40 ppb level. The major SOA precursors from heated cooking oils were mono-unsaturated fat and omega-6 fatty acids. It was also noticed that animal fats content (pork and chicken fat) are also abundant in mono-unsaturated fat and omega-6 fatty acids, thus gas-phase emissions from cooking animal fat was suggested to be efficient in producing SOA.

Another chamber study with proton-transfer-reaction time-of-flight mass spectrometer (PTR-ToF-MS) showed gaseous emissions from different cooking activities [27]. During shallow *frying of meat* (based on calculations of emission factors: chicken - 3 mg kg^{-1} ; beef - 42 mg kg^{-1})

the dominant gaseous compounds were aldehydes (from 30% to 60%, depending on type of oil used) compared to *vegetables frying* when lower aldehyde emissions were observed as a result of low temperatures (150 °C). Monoterpene emissions (3 mg kg⁻¹) during vegetable frying were attributed to emissions from condiments (oregano, basil). *Deep frying* of potato resulted in alcohol and aldehyde emissions. *Charbroiling of beef and chicken* resulted in saturated aldehydes emissions as ground beef mostly consists saturated or singly unsaturated fatty acids. Charbroiling of burger patties had contributed of octanal emissions (7 mg kg⁻¹).

Vegetable boiling was emitting acetaldehyde when treating zucchini (0.3 mg kg⁻¹) and explained to originate from drought stress conditions during the growth, mechanical stress after the harvest or storage under low oxygen conditions. Boiling carrots was emitting acetaldehyde (0.5 mg kg⁻¹), and sesquiterpenes (0.3 mg kg⁻¹) pointed out that they could be formed during root development in the plants. The VOCs emissions from *heated vegetable oils* (canola, sunflower, olive oil) were dominated by aldehydes: acrolein (C₃H₄O), malondialdehyde (C₃H₄O₂), 2,4-heptadienal (C₇H₁₀O), nonanal (C₉H₁₈O), hexanal (C₆H₁₂O), 2-heptenal (C₇H₁₂O), 2,4-decadienal (C₁₀H₁₆O), heptanal (C₇H₁₄O), octanal (C₈H₁₆O), -decenal (C₁₀H₁₈O), and 2-undecenal (C₁₁H₂₀O). These aldehydes are not present in the oils, but chemically produced upon heating.

During *shallow and deep frying* using different oils and from charbroiling in all emissions hexanal and nonanal have been observed. In the ambient conditions hexanal and nonanal have relatively long atmospheric lifetimes (18 h for hexanal and 15 h for nonanal) which means that it is sufficient time for formation of secondary organic aerosols in the presence of ozone.

In epidemiological studies the exposure to outdoor fine particulate matter PM_{2.5} have been linked to cardiopulmonary diseases and increased mortality. The large particle fraction from cooking-related activities are emitted in the ultrafine and fine modes of different chemical nature, this may lead to a deleterious effect on the respiratory system. Additionally, cooking fumes were found to contain carcinogenic (polycyclic aromatic hydrocarbons; aldehydes - acetaldehyde is classified by the IARC as «possibly carcinogenic») and mutagenic compounds causing irritation of the airway mucosa, pneumonia and have a risk of respiratory cancer [35].

2.1.2 Emissions from combustion sources

Candles are known to be used with religious and aesthetic purposes. For a candle to burn, the heat of the flame is used to light the candle's wick which instantly turns the wax (as it has low melting point) into a hot liquid. The liquid fuel is then drawn up the wick by capillary forces and vaporizes under high temperature, breaking down hydrocarbon molecules into hydrogen and

carbon. These vaporized molecules are oxidized to ignite and form a constant flame. In the oxygen-rich blue zone, the hydrogen is the first to separate and react with the oxygen to form water vapor. In fuel rich regions of the flame, large quantities of soot particles form. The yellow incandescent light making up the majority of the flame is the result of soot oxidation in the flame. During steady burn almost all soot formed in the flame can be oxidised, resulting in low elemental carbon emissions. The candle flame could be affected by air movements (for instance, by window opening) and the flame would flicker resulting in incomplete combustion of the soot, soot particles will coagulate with newly formed ultrafine particles - sooting burn mode [54]. The amount of soot produced can vary greatly, for instance, in a study by [67] elemental carbon emission rates varied from <40 to $3,370 \mu\text{g g}^{-1}$ in sooting behaviour.



Fig. 2.1 Candle burning [83]

[54] have been assessing physico-chemical composition of two types of candles (candle I: stearin-based; candle II: mix of stearin and paraffin) in a chamber study. Generally, both candles have generated high number concentration of ultrafine (<100 nm, peaks in GMD of 20–30 nm) particles during steady burn ($1100\ 000$ particles cm^{-3} for candle I and $5.1 \cdot 10^6$ particles cm^{-3} for candle II).

During sooting burn mode less ultrafine particles were emitted ($890\ 000$ and $270\ 000$ particles cm^{-3} for candle I and II) in comparison to steady burn, due to coagulation of ultrafine particles with soot particles. [50] have reported size distribution have been dominated by ultrafine particles of around 30 nm in diameter during steady burn of a single paraffin wax candle.

Candles that are extinguished by blowing out the candle, produce more soot than those extinguished by cutting off the tip of the wick. Cutting the wick eliminates the emissions produced by a smouldering candle.

Besides that, soot is primarily composed of elemental carbon, the candle soot may include metals, inorganics, and volatile organic compounds (benzene, toluene, acetaldehyde, formaldehyde, acrolein, naphthalene, terpenes. Metal (such as, lead, zinc, tin) was originally put in wicks to keep the wick standing straight when the surrounding wax begins to melt. In 1974, once it was shown that burning lead-wick candles resulted in increased lead concentrations in indoor air, it was voluntarily agreement US candle manufacturing industry to cease production of lead-containing candles [50]. However, lead wick candles can still be found on the market throughout the world. Zinc is another alternative since it provides the desired amount of stiffness, burns off readily with the rest of the wick, and does not have the same toxic effects as lead. Zinc

is an essential element for human health, inhaling large amounts of zinc (as zinc dust or fumes from smelting or welding) over a short period of time can cause a metal fume fever disease [50]. Phosphates and alkali nitrates were also found in candle emissions which were added as flame retardants to the wick [54]. Emitted volatile organic compounds in presence of oxidants may lead to secondary particle formation. Candle soot has also shown similarities in morphology to diesel soot, but with low PAH emissions.

Scented candles include various fragrance oils which are unsaturated hydrocarbons, thus has higher carbon-to-hydrogen ratio compared to paraffins (which are saturated hydrocarbons). Therefore, waxes that have more fragrances in scented candles produce more soot.

Eventually, when soot builds up in air it may deposit onto surfaces on one of the such scenarios: (1) the particle may randomly collide with a surface; (2) particles can gain enough mass to become subject to gravitational settling; (3) the particles could be attracted to electrically charged surfaces - freezers, vertical plastic blinds, television sets, computers.

Airborne soot particles can potentially penetrate to the human lower respiratory tract [50]. [50] subjected candle flame particles to relative humidity comparable to that in the human respiratory tract, and found that candle flame particles grew in size. It was supposed that found water-soluble combustion particles (ammonium phosphate and alkali nitrates) may have growth potential in the respiratory tract. White candles had a 20% growth potential larger than scented candles.

Incense produces particulate matter of 0.1 to 0.7 μm , and elevates concentrations of carbon monoxide and benzene. Incense also contains trace amounts of chemicals suspected of causing skin irritation, and exposure to incense has been linked with several illnesses, including cancer, asthma, and contact dermatitis [50].

2.1.3 Emissions from cleaning and household products

Cleaning activities such as sweeping, hovering, dusting (resuspension from carpets), operations with washing powders tend to emit particles in coarse mode indoors [29]. Using of common household products such as cleaning products, air fresheners, detergents, perfumed or scented products, polishes, can cause an increase in the indoor airborne concentrations from primary emissions of *gaseous* [68], *particulate species* and drive various chemical reactions (ozone-terpene chemistry) forming *secondary organic aerosols* in the ultrafine range. [61] investigated the influence of the use of commercial cleaning agents on a pine oil basis on the exposure in a living room. During the activities, the PNC rose from initially 2000 particles cm^{-3} to a maximum of 190 000 particles cm^{-3} and the PM_{2.5} contents increased from 5 to 38 $\mu\text{g m}^{-3}$. The

increase in number and mass concentrations was explained by the formation of secondary organic aerosols (SOA) or particle growth condensing on a pre-existing particle causing their mass and size to increase.

Formation of secondary organic aerosols (SOA) [26; 32] takes place when ozone reacts with terpenes and forms oxygenated organic compounds. Products of the reaction (organic acids and aldehydes) having low vapor pressure can condense from the gas phase to airborne particulate matter. Previously reported size range of SOA was ~15 nm to 700 nm [75].

Terpenes, which are volatile organic compound (VOC), are widely used in cleaning sprays, perfumes, air fresheners, candles, cosmetics, either as solvents or to provide a distinctive scent. The most common ingredients in terpene cleaners are D-limonene, a derivative from orange peels; alpha and beta pinene from wood turpentine; camphene; 3-carene; α -terpinene [42]. Although terpenes themselves are not considered toxic, but being oxidized (in the presence of ozone, hydroxyl or nitrate radicals) indoors, they are resulted in producing other volatile organic compounds (i.e. formaldehyde which is known as "carcinogenic to humans" by IARC) and secondary organic aerosols.

Indoor ozone mostly originates from outdoor environment [28] but could also be generated from printers and photocopiers [30]. Hydroxyl radicals and nitrate radicals are other oxidants that may be present indoors at sufficient concentrations to produce secondary products. Hydroxyl radicals are produced during reactions of ozone with a various VOCs [28], and indoor nitrate radicals result from the reaction between ozone and nitrogen dioxide.

For instance, the lifetimes of d-limonene and α -pinene in the normal ozone concentration (50 ppb) are 0.75 and 1.8 h, respectively [76]. The residence time of air indoors before being replaced by ventilation is enough for secondary-product formation.

Some studies showed that exposures from various types of cleaning products - for furniture, carpets, rugs, curtains, glass and mirror cleaning sprays, ironing sprays, air fresheners, detergents, perfumed or scented products, polishes, have been linked to increase in asthma and other respiratory diseases [34]. Indoors, they are resulted in producing other volatile organic compounds (i.e. formaldehyde, which is known to be "carcinogenic to humans") [IARC] and secondary organic aerosols.

2.1.4 Emissions from other indoor sources

Particulate matter may also be emitted from photocopiers, laser printers in fine particle mode or came from housekeeping – hovering, sweeping, ironing [9], human movements, or to be resuspended from clothes [43] in a coarse mode.

2.2 RESUSPENSION

Particles that have been deposited on the surfaces may become resuspended through activities indoors (walking, housekeeping) and re-entrained into the air [53]. Resuspension may be important pathway for allergens exposure [69] and also for semi-volatile species such as phthalate esters [36]. [62] quantified effective mass emission rates of PM_{2.5} and PM₅ in one house associated with prescribed household activities: folding blankets, folding clothes, dry dusting, making a bed, dancing, vacuuming, walking, and sitting on upholstered furniture.

2.3 DEPOSITION

The *deposition loss rate coefficient* is defined as the number of particles depositing on the total surface available per unit of time divided by the total number of particles in the air, and given in units of h⁻¹ [59].

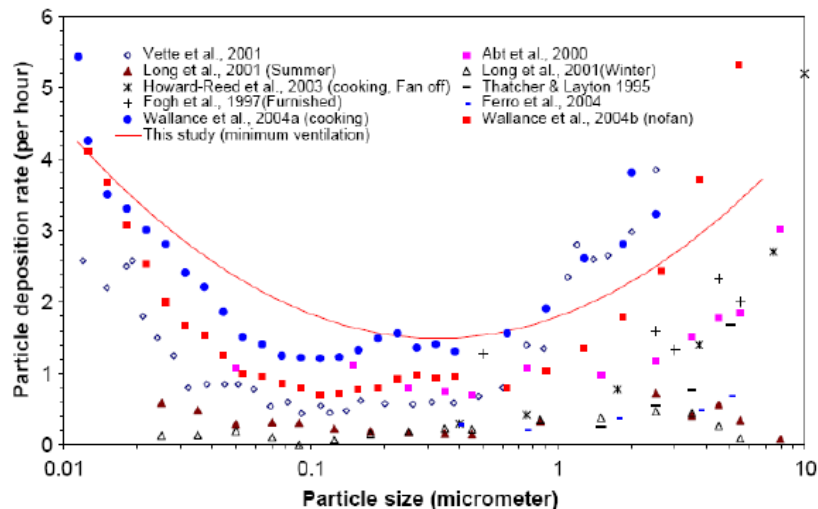


Fig. 2.2. Relationship between the deposition loss rate coefficient and particle sizes measured in real houses [70]

Particle deposition on indoor surfaces is size-dependent and is governed by deposition mechanisms for small particles (<0.1 μm) by diffusion towards the surfaces, and for larger particles (>1 μm) by gravitational sedimentation [53]. For particle sizes between 0.1 and 1 μm , both mechanisms are weak and the particles can stay airborne for significantly long time [59], Figure 2.1. The studies have concluded that deposition process is strongly dependent on particle size, reaching a minimum for particle diameters in the range 0.1 to 0.3 μm [53].

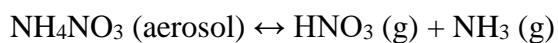
In addition, the presence of airflows induced by convection currents or the action of fans can increase particle transport towards the surface and thus the deposition. Deposition is also dependent on the area of the surface: the larger the surface area, the higher the probability of

particle deposition (furnished rooms, with lots of surface area, will have higher deposition rate than bare rooms). It also dependent on surface characteristics: sticky surfaces resulting in higher deposition than smooth ones. Additional factors affecting particle deposition are: the presence of surface charge, which increases the deposition rate; temperature gradient, which results in convective currents and thermophoretic deposition; and room volume [53].

2.4 PHASE CHANGE

During outdoor-to-indoors transport, particles undergo to phase-change processes, in which chemical species change their state from gas phase to condensed phase, or vice versa [69]. Some of outdoor origin particles are considered to be non-volatile, which means that their vapour pressure is low and they are always found in the particle phase. The other group of compounds is semi-volatile which may exist in a gas, liquid, or solid particle phase depending on temperature and relative humidity parameters [71].

Ammonium nitrate, NH_4NO_3 , is a major component of atmospheric aerosol species. Ammonium nitrate is semi-volatile thus the gas-to-particle partitioning is highly susceptible to changes in temperature, relative humidity and the gas phase concentrations of ammonia and nitric acid as outdoor air during transport indoors [21; 19]. During the winter period, the nitrate aerosol moved from a cooler outdoor to a warmer indoor environment, shifting the equilibrium of the nitrate formation reaction, towards the gas phase:



Dissociation of NH_4NO_3 indoors is not only driven by temperature and relative humidity, but also by the interaction of the particles with surfaces such as walls, furniture, ceilings and other structures. The gases diffuse toward and absorb to surfaces inside the house, further driving the equilibrium toward the gas phase [19].

Sulphate is not volatile compound and has been shown to have little dependence on temperature [72; 73]. It can be also considered to be undergo size dependent penetration from outdoor environment.

2.5 PENETRATION OF OUTDOOR PARTICLES

Indoor PM concentration is affected by outdoor-to-indoor transport of outdoor origin particles. For instance, outdoor particles from natural sources (Kampa et al. 2007), biomass burning, vehicle emissions [16; 47; 17; 77; 78; 79; 80] can be transported indoors where humans

spend most of their time [55] mixing with indoor pollutants generated by cooking, candle or incense burning, using of the household products and emitted from indoor materials (Rackes and Waring, 2015). The fraction of particles in the infiltration air that passes through open windows or doors or through any cracks, gaps or holes to the building envelope called *penetration factor*, P [53]. Penetration through the building envelope is size dependent with the highest penetration accumulation mode particles [69] since these particles have a rather low diffusivity and are too small to be affected by sedimentation or impaction.

The study showed [81] that penetration efficiency (2-90 %) through the horizontal slit (0.508 mm high, 102 mm deep, and 433 mm wide) for 2 μm particles was increasing 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 dropped rapidly, reaching almost zero in the 0.01 μm range. When windows or doors of a building are open, they provide the main penetration route for the all size ranges particles, giving the large air-exchange rate, and the relative importance of penetration through the building envelope becomes insignificant.

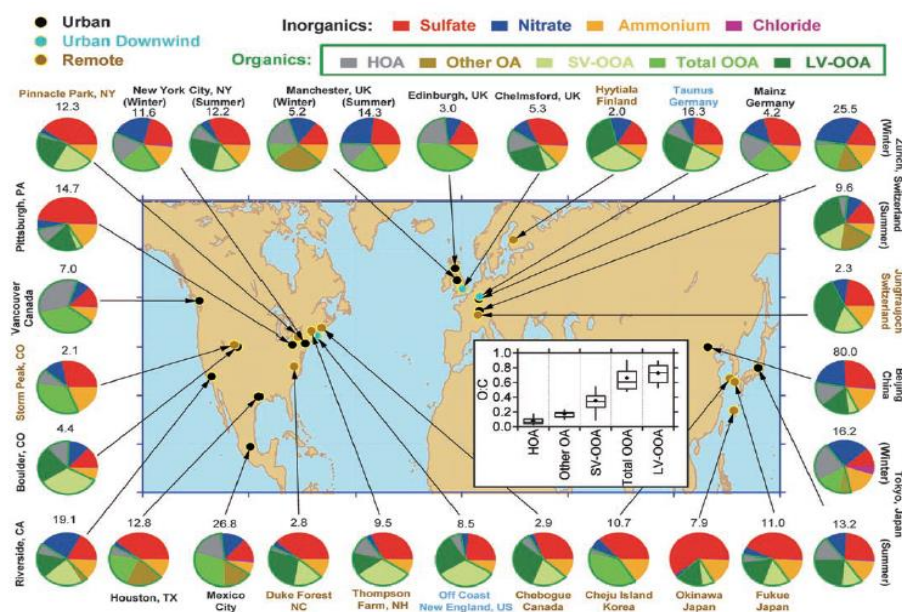


Fig. 2.3. Total mass concentration ($\mu\text{g m}^{-3}$) of nonrefractory inorganic species and organic components in submicrometer aerosols (measured with the Aerosol Mass Spectrometer in the Northern Hemisphere), [74]

Outdoor environment consists of a complex combination of gases and particulate matter (PM). PM is emitted directly into the atmosphere (primary PM) or formed minutes/hours after the first gaseous emissions in the atmosphere through gas-to-particle conversion (secondary PM) [33; 66]. In particular, fine particulate matter (PM_{2.5}) impacts human health, visibility, the ecosystem, the climate, and these PM effects are largely dependent on the aerosol properties, including the

number concentration, size, and chemical composition. The chemical compositions of fine PM usually include sulphate, nitrate, ammonium, inorganic ions, elemental carbon (EC), organic carbon (OC), and metals [77]. At different locations, times, meteorological conditions the relative abundance of different chemical components can vary (Figure 2.3). Particles, which are emitted directly from vehicles, like the particles from other combustion sources are small with the majority of them belonging to the ultra-fine size range ($< 0.1 \mu\text{m}$). Nuclei particles have relatively short lifetimes in the atmosphere because of their high number concentration near emission source these small particles coagulate rapidly with each other and end up in the accumulation mode [49].

The coarse-particles ($\text{PM} > 2.5 \mu\text{m}$) are emitted from wind-blown dust, large salt particles from sea spray, and mechanically generated anthropogenic particles such as those from agriculture and surface mining [49]. Because of their large size, the coarse particles are heavy enough that gravitational settling removes them quickly to prevent the build-up of large number densities [74]. While particles larger than $1 \mu\text{m}$ are removed by gravitational settling, and particles with diameters $< 0.1 \mu\text{m}$ are able to diffuse through the air, and thus end up on surfaces, particles with diameters in the range of 0.1 to $1 \mu\text{m}$ are not effectively removed by either mechanism. As the name suggests, particles accumulate in this mode because removal mechanisms are weak. Particles can be removed by rainout or washout, but they coagulate too slowly to reach the coarse-particle mode.

2.6 INDOOR/OUTDOOR (I/O) RELATIONSHIP

The particle concentration indoors is affected by a number of important parameters which are summarized in mass balance equation:

$$\frac{dC_{in}}{dt} = aPC_{out} + \frac{S}{V} - aC_{in} - kC_{in} \quad (2.1)$$

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

t - time,

a - air exchange rate due to infiltration (h^{-1}),

P - particle penetration factor,

k - decay 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* is used to show the difference between indoor and outdoor particle concentrations, and depends on infiltration factor, indoor emissions, indoor losses by filtration or deposition.

$$\frac{I}{O} \text{ ratio} = \frac{C_{in}}{C_{out}} \quad (2.2)$$

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

Infiltration factor represents the equilibrium fraction of outdoor origin particles that penetrates indoors and remains suspended. In the absence of indoor sources for suitably long time the indoor/outdoor ratio is equivalent to the infiltration factor [59].

$$F_{in} = \frac{aP}{a + K} \quad (2.3)$$

In the absence of indoor sources the infiltration factor depends on air exchange rate, deposition rate and penetration factor in naturally ventilated houses [61]. [53] found I/O ratios for PM_{2.5} with no indoor sources and no human activity were from 0.54 to 1.08 and when indoor emission sources were present, I/O ratios ranged from 1 to 2.4 in naturally ventilated buildings.

3. METHODS

3.1 TIME-OF-FLIGHT AEROSOL MASS SPECTROMETER (ToF-AMS)

ToF-AMS was used to measure chemically-resolved mass loadings and size distributions of inorganic and organic species in the range of 50-500 nm. The AMS consists of three main parts: an aerosol inlet, a particle sizing chamber, and a particle composition detection section. The different sections are separated by small apertures and differentially pumped reducing the sampled gas flow. Aerosols are sampled through 100 μm critical orifice into an aerodynamic lens which focuses particles of different sizes into tightly collimated particle beam by 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.

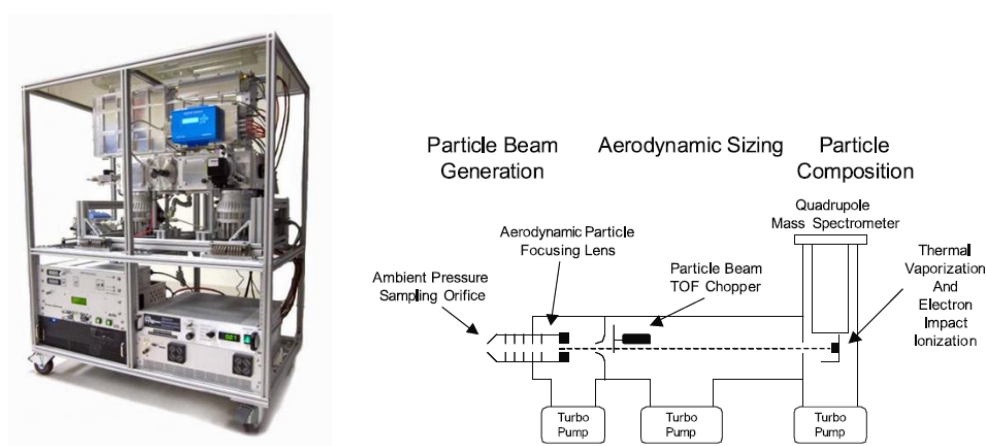


Fig. 3.1. Schematics of an Aerodyne Aerosol Mass Spectrometer [20]

Size-dependent particle velocities created in a response to the gas expansion, caused by the pressure drop upon exiting from the lens, provide a means of obtaining the aerodynamic diameter by measuring particle time of flight. From there, particles are impacted on a heated surface (~ 600 $^{\circ}\text{C}$) where nonrefractory aerosol components flash-evaporate quickly (within 50–100 μs). The resulting vapor molecules are ionized by 70 eV electrons (tungsten filament as the electron source) which is enough energy to cause extensive fragmentation and do not significantly affect the samples. The electrons, colliding with vaporized molecules, pull out the electrons from the molecular electron shells and convert them into positive ions, to analyse via mass spectrometry according to their mass-to-charge ratio (m/z). For the detection of the compounds, everything has to occur under high vacuum conditions, otherwise electrons will charge the air molecules rapidly, and ions formed from compound of interest will turn too quickly into neutral molecules. Only organic species and most nitrate and sulphate salts are detected, whereas crustal material, sea salt, and black carbon are not [20; 22]

3.2 SCANNING MOBILITY PARTICLE SIZER (SMPS)

SMPS was used to detect particle number concentration and size distribution of mobility diameter between 13 and 615 nm during the measuring campaign. SMPS consists from a bipolar charger, differential mobility analyser (DMA, 3081) and condensation particle counter (CPC, 3775). The technique is based on the ability of charged particles to traverse in the electrical field according to their electrical mobility.

In the bipolar charger particles are acquiring equilibrium charge distribution and are introduced to the DMA where particles are driven by the sheath flow through the electric field created between inner collector rod with negative potential and outer electrically grounded one.

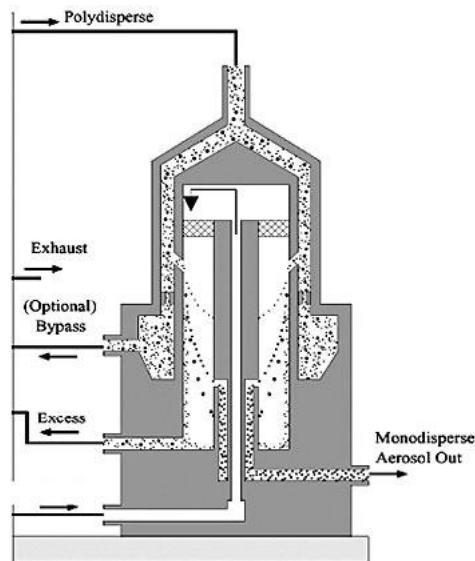


Fig.3.2. Principal scheme of Differential Mobility Analyser [24]

Positively charged particles are attracted by central collector rod according to their electrical mobility. Particles with a narrow range of electrical mobility exit with the monodisperse air flow and move towards CPC. The remaining particles are removed from the DMA via excess air.

When entering to the CPC particles are saturated with the alcohol vapor (butanol-based CPC) with subsequent cooling in a condenser. The alcohol condenses on particles in the sample flow creating droplets large enough to be efficiently detected by optics [24]. As droplets exit the condenser, they pass through a thin ribbon of laser light. Light scattered by these droplets is collected by optics and focused onto a photodetector. The photodetector converts the light signal to an electrical pulse, which is recorded as a particle count.

3.3 SAMPLING

3.3.1 Sampling site

Indoor and outdoor measurements were conducted during one-month campaign in December, 2016 – January, 2017 in an occupied residence in Malmo, Sweden. The ground-floor apartment was naturally ventilated (total space area of 117 m², 275 m³), had four rooms – three bedrooms, living room, two bathrooms, dining room and kitchen with the kitchen fan installed.

The apartment was located in a three-storey concrete building, enclosed from other areas by green zone. As the sources of ambient air pollution could be accounted: use of fireplaces in neighbour's apartment, adjacent fast food restaurants and local traffic within a few blocks.

3.3.2 Sampling methodology

Two parallel sampling lines (indoor and outdoor) with automatically switching valve were used. The valve alternated the measurements between indoor and outdoor for 20 and 10 minutes, respectively. Stainless steel tubes were mounted at the ground floor level near the window frame leading to the basement area (length 3,27 m, diameter 5 mm) (Figure 3.3). Indoor sampling line was heated and insulated, additional carrier flow was used to lower the residence time and ensure the same temperature. After the switching valve, sampled aerosol passed through in the Silica dryer (Perma-Pure, length 47 cm, RH < 30%) and then split into lines which go directly to the Time-of-flight Aerosol Mass Spectrometer (ToF-AMS) and to the Scanning Mobility Particle Sizer (SMPS).



Fig. 3.3. Sampling site

The mass of the chemically resolved nonrefractory submicron particles between 50 and 500 nm was detected by AMS (scanning time - 30 s). The SMPS was measured particle number and size distributions between 13 and 615 nm mobility diameter (scanning time - 3.5 minutes).

The total inlet residence time of the particles in lines was 4 seconds to the Aerosol Mass Spectrometer (AMS) and 27 seconds for Scanning Mobility Particle Sizer (SMPS). The scans between switches were not considered as they represented joint indoor and outdoor scans. Particle losses were calculated due to diffusion of the submicrometer particles in the long sampling lines. Correction for diffusional losses were done only for the SMPS data. While to account of lost fractions of particle chemical species for Time-of-Flight Aerosol Mass Spectrometer (ToF-AMS) more detailed calculations are needed since determined species could possibly condensate in the stainless-steel tubes.

Sampling was done during winter period, this would give better understanding about the particle loadings indoors with reduced air exchange rate, as well as accessing the differences in particle number and mass concentrations inside and outside of the residence. At the beginning of the measuring campaign the occupants were getting logbook to keep track of the usual indoor activities, such as frying, boiling, kitchen fan operation, baking, toasting, use of the microwave, candle burning, using of household products, hoovering, sweeping, ironing, printing operations, windows opening (indoor activities logbook, Appendix B) and the exact time periods when the activities took place.

Within the measurement campaign the periods of everyday indoor activities were always alternating with more vigorous indoor events (measurements captured the period of holidays). Additionally, instruments were not continuously running all the time due to the flow check-ups or other technical reasons. The chosen for analysis 7-day period covers periods of intensive and regular indoor events to look on the real situation in the apartment for the assessment of the particle loading during this time.

3.4 DATA ANALYSIS

The comparison between indoor and outdoor loadings was done in order to understand contribution of each indoor and outdoor particle origin to the indoor loadings and to find the best way for personal exposure assessment to particulate matter indoors for studying health-related effects. Data analysis was done based on particle number size distribution, number concentration (data from SMPS) and mass concentration of non-refractory species (data from AMS) for the following periods:

- (a) *Total monitoring period*, which is defined as the entire monitoring duration and includes periods of occupancy and non-occupancy. The analysis of total monitoring period may underestimate the concentrations for exposure assessment since include the non-occupancy time.
- (b) *Occupancy period* is defined as the time when at least one person is present in indoor environment. Data from this period was studied for better estimates of particle concentrations and characteristics of exposure assessment to particulate matter indoors.
- (c) *Non-occupancy period* is the time when no one is present in the residence. This data was used for studying influence of the outdoor particles upon entering indoors [58].

4. RESULTS AND DISCUSSION

4.1 PARTICLE NUMBER CONCENTRATION (PNC)

Calculated values of particle number concentration (PNC) for total monitoring period, occupancy, non-occupancy periods inside and outside of the apartment are presented in Table 4.1. Average particle number concentration during whole monitoring period indoors (which comprise from occupancy and non-occupancy periods) was higher in comparison to outdoor concentration. Average concentration during occupancy time accounting all indoor events during the whole analysed period have resulted in average value of 6 100 particles cm^{-3} , while in the absence of the occupants and thus activities concentration was equal to 2 800 particles cm^{-3} . The difference in PNC between these two periods could be explained by the contribution from indoor sources generated during occupancy time (reaching maximum concentration of 234 400 particles cm^{-3} during these activities) and possibly, due to low air exchange rates in the naturally ventilated apartment during winter period (i.e. people do not open windows so frequently in winter compared to other seasons). The characteristic active indoor sources include various types of cooking - frying, deep frying, baking, toasting, breakfast preparation, candle burning, ironing of the clothes. [5] and [2] reported cooking activities and candle burning as the main contributors to the particle loadings indoors. Frequent burning of candles which may last several hours during the winter months are not unusual in Scandinavia [5].

Table 4.1. PNC summary statistics during one-week period (SMPS data)

		Total monitoring period ¹		Occupancy period		Non-occupancy period	
		Indoors	Outdoors	Indoors	Outdoors	Indoors	Outdoors
Average	particle/ cm^3	5900	2088	6100	2040	2800	3174
STD ²		23300	1213	23800	1208	1800	718
I/O ratio		2.8		3.0		0.9	
Median		1200	1924	1200	1875	2400	3111
Maximum		234400	14270	234400	14270	9600	5017
Minimum		200	326	200	326	1200	1946

¹ Data for the whole analysed period.

² Standard deviation.

Non-occupancy period indoors showed lower number particle concentrations in comparison to respective outdoor values. Indoors, period without operation of indoor emission sources reflects penetration of outdoor origin particles which is supposed to be size-dependent [69]. Important to note, that some periods within non-occupancy time were influenced by elevated concentration generated previously from indoor activities, e.g. - occupants were cooking before leaving apartment. If the influence of that remaining concentration from previous indoor events was

excluded the values for non-occupancy period would be lower. In context, [5] found higher PNC during occupancy time in 56 Danish homes for the particle size range of 10-300 nm (in particles cm^{-3} : average 35 500, STD 52 300, median 13 100, max 304 400) in comparison to non-occupancy (in particles cm^{-3} : average 8200, STD 9900, median 6200, max 71200) indicating rather high indoor particle loading during this study. [58] have also found higher PNC in naturally ventilated apartment (particle size range of 15-700 nm) for occupancy time (in particles cm^{-3} : average 12 000, STD 30 000, median 1900, max 246 000, min 700) compared to values in the present thesis. The differences in PNC particle loadings reported by [5], [58] and values found for the present apartment could be explained by more frequent indoor activities in the previous studies, differences in air exchange rates, types of ventilation system, building characteristics, volumes of the homes, and variations of outdoor loadings. Additionally, the occupants of the apartment studied by [58] were not always using kitchen exhaust fan during cooking, enhancing accumulation of pollutants indoors.

The particle loadings outside of the apartment comprised from traffic emissions, local sources, such as emissions from fireplaces used by neighbours, and from adjacent fast food restaurants. The variations of both indoor and outdoor concentrations could be seen in the Figure 4.1.

Indoor-to-outdoor ratio (I/O) turned to be higher during occupancy time 3.0, while during period with no indoor sources and no human activity indoor-to-outdoor ratio was equal to 0.9. Lower value of the ratio during non-occupancy time can be also explained by protective role of the building i.e. not all particles from outside penetrate from outside. [53] summarized found I/O ratios in few studies for PM_{2.5} from 0.54 to 1.08 during non-occupancy time, and when indoor sources were present I/O ratios ranged from 1 to 2.4 in naturally ventilated homes. The higher I/O ratio for occupancy period in the studied apartment than in [53], was due to higher contribution from indoor activities, different air exchange rates, particle size range measured, and smaller volume of the present apartment compared to the homes studied by [53].

Figure 4.1 represents indoor particle number concentration inside and outside of the apartment one-week period. The duration of the known indoor activities was marked on the graph from the activity logbook. The graph shows increasing number concentration indoors when various indoor sources were in operation. Type of activity (cooking, candle burning, ironing), its duration, way of performing indoor events may result in different number particle concentrations. For instance, candle which was burning almost for the same duration of 3.5 and 3 hours (16th and 21nd of December, Figure 4.1) have resulted in different number particle concentrations of 23 000 particles cm^{-3} and 8 200 particles cm^{-3} (in both cases candles of the same type were burned). The variations in the particle concentrations may be due to the different candle burning mode – the

candle flame could be affected by air movements (for instance, by window or entrance door opening, human movements) causing flame to flicker and burn in the sooting mode: some soot particles are able to escape without being oxidized, decreasing number concentration of ultrafine particles due to coagulation of soot mode particles with newly formed ultrafine particles [54].

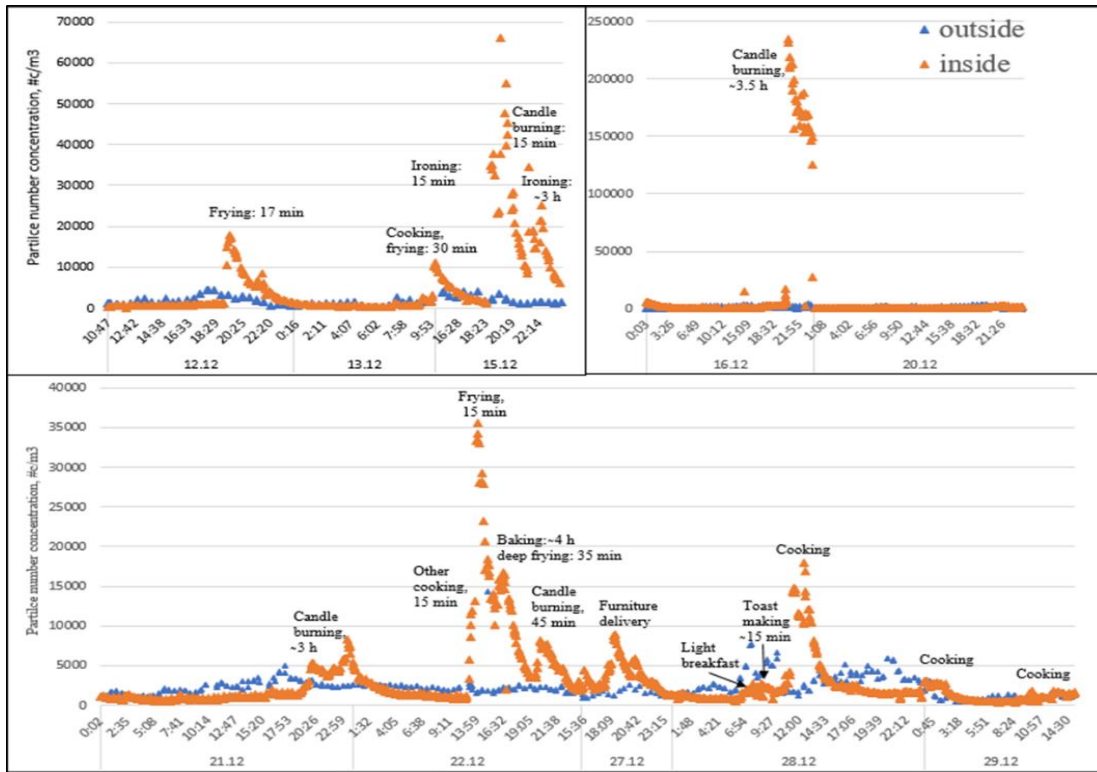


Fig. 4.1. Particle number concentration inside and outside of the apartment during one-week period (SMPS data)

[9] have reported particle number concentrations (measured range of 10-400 nm) from 2 candles which were simultaneously burning for 40 minutes and showed 10 000 particles cm^{-3} in the living room, and produced levels $> 200\,000$ particles cm^{-3} close to SMPS inlet. The variations in number particle emissions between [9] and value found in this thesis (highest peak of 23 000 particles cm^{-3}) may be due to the longer time of candle burning in the present apartment (up to 3.5 h), candle burning modes, type of candles used (scented candles tend to produce more soot particles compared to white candles), bigger volume of the apartment in [9] resulted in more efficient dispersion of the pollutants indoors. [54] in the chamber study (22 m^3) were assessing candle emissions in the measured particle size range of 16–1000 nm, and found 1140 000 particles cm^{-3} (I: for stearin-based candle) and 510 000 particles cm^{-3} (II: for candle from stearin and paraffin mix) during steady burn. During sooting burn it was 890 000 particles cm^{-3} and 270 000 particles cm^{-3} for candle I and II. The number particle concentration measured in the chamber may differ from the real indoor environment where particles of different origin are present (newly

formed particles may coagulate faster with already existing ones) and other factors - likelihood of more frequent air movements in the apartment causing candle to burn in the sooting mode.

Cooking emissions resulted in peak concentrations for frying 35 500 particles cm^{-3} , baking 16 000 particles cm^{-3} , toast making 2 600 particles cm^{-3} , breakfast preparation 2 300 particles cm^{-3} , during ironing it was 34 400 particles cm^{-3} . [2] have found (measured particle range of 10-300 nm) for frying 14 000 particles cm^{-3} , for baking 230 000 particles cm^{-3} , for toasting 160 000 particles cm^{-3} .

4.1.1 Number size distribution during periods of monitoring

Particle number and mass size distributions indoors and outdoors were monitored in the size range of 13–615 nm. Average *number size distributions* were plotted for total monitoring, occupancy/non-occupancy periods inside the residence and respective outside concentrations in Figure 4.2. The characteristic differences between these periods were the following:

(1) The total monitoring period and occupancy time have had the identical size distributions since most of the time the occupants were present in the apartment (90 % of the total monitoring time).

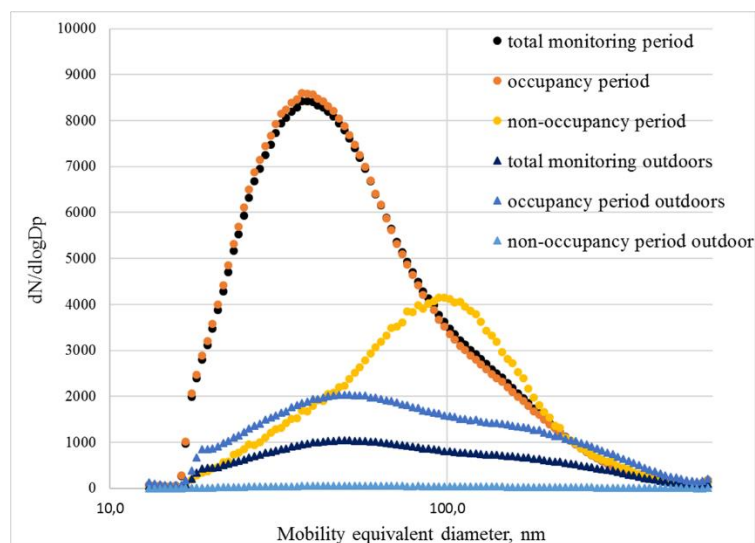


Fig. 4.2. Number size distribution during total monitoring, occupancy and non-occupancy periods inside and outside of the residence

(2) Occupancy time have accounted all indoor activities which were constantly occurring within this analysed period, and the influence of the outdoor origin particles. Both number size distributions during total and occupancy periods showed to be distributed in the ultra-fine (<100 nm) particle size range (84% of total PNC), with the mode about 40 nm. Such size distribution within occupancy time is important in relation to human health since UFP have higher deposition

rates in the lower respiratory tract than larger ones [49]. Particle levels indoors during the period of occupancy were higher compared to outdoor ones more than in 4 times.

(3) Size distribution within non-occupancy period should reflect influence of outdoor particles. But this period was affected by concentration generated previously from indoor activities (cooking before leaving apartment), showing particle growth due to coagulation and condensation of other components on particle surfaces.

4.1.2 Number size distribution from indoor sources

Number size distribution during occupancy period indoors showed to be distributed in UFP range which may indicate that the majority of the indoor sources could be also distributed in the same particle range.

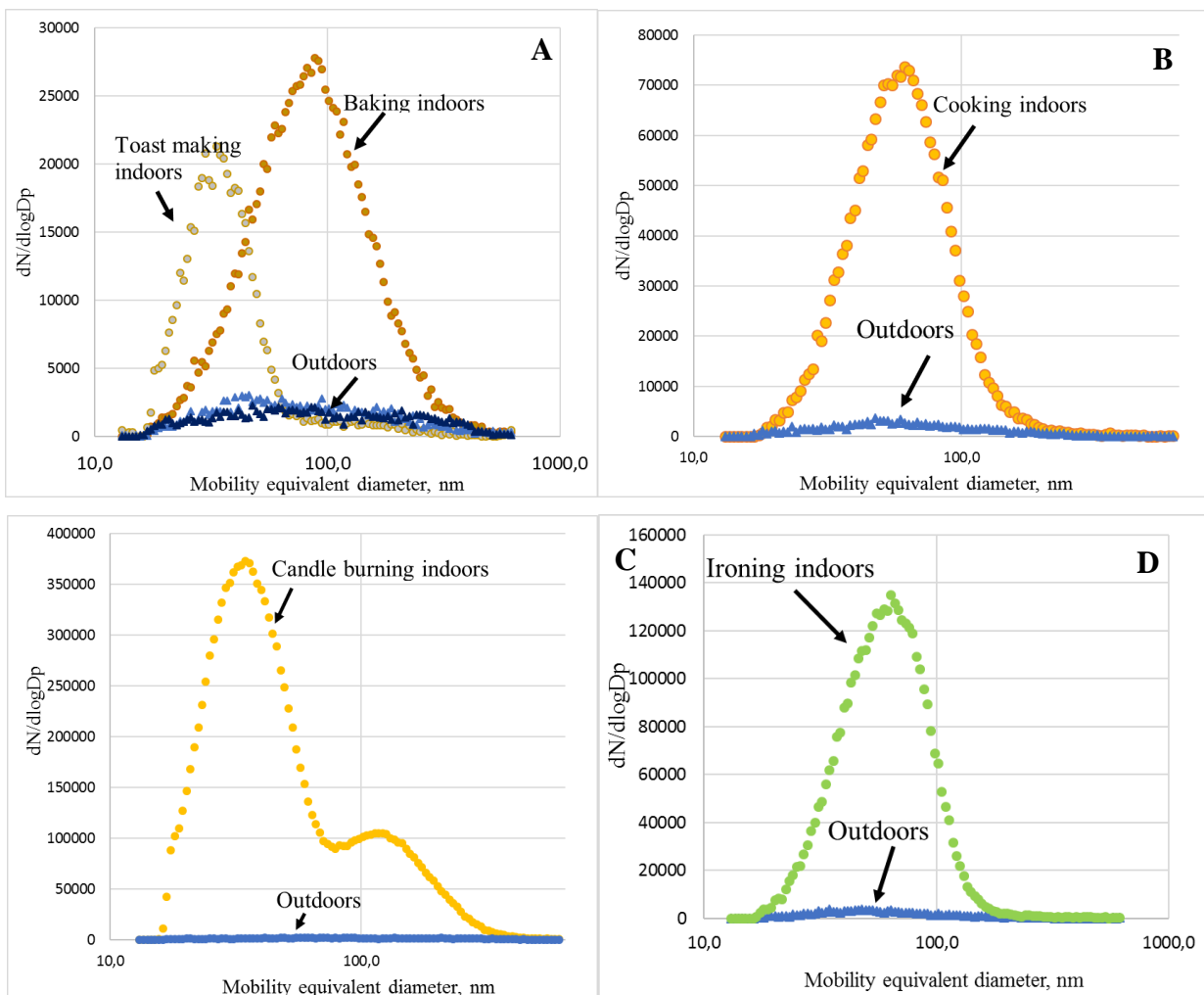


Fig. 4.3. Particle number size distribution indoors and outdoors during: A, B) cooking activities, C) candle burning, D) ironing

In order to confirm this, in the Figure 4.3 it was plotted particle number size distributions of individual indoor activities, such as, baking, toast making, cooking, candle burning, ironing, and respective outdoor concentrations. The outdoor concentrations were put at the same time when each of these indoor activities took place. Each graph shows one scan (3.5 minutes) on the basis of SMPS data. As one can see, when specific indoor activities were in operation, majority of them were distributed in UFP mode, and have substantially exceeded outdoor concentration.

Number size distribution during cooking resulted in PNC of 72 900 particles cm^{-3} with the mode around 85 nm, for baking 27 500 particles cm^{-3} and the mode ~95 nm, when making toast 19 200 particles cm^{-3} and the mode ~35 nm, for ironing 135 000 particles cm^{-3} and the mode ~60 nm. Particle size distribution during candle burning showed bimodal size distribution (Figure 4.3D), with the particle number of the first peak of 371 200 particles cm^{-3} with the mode ~35 nm and second mode ~130 nm. The second mode may indicate on the sooting burn mode [54]. The flickering of the candle flame, as the result of horizontal air movements (for instance, by window or entrance door opening, human movements), some soot particles are able to escape without being oxidized and coagulate with newly formed ultrafine particles and already existing particles indoors.

4.1 PARTICLE MASS CONCENTRATION (PMC)

Table 4.2 summarises averaged particle mass concentration of organic, nitrate, sulphate, ammonium and chloride particles measured by Aerosol Mass Spectrometer, presented for the whole monitoring, occupancy and non-occupancy periods. Overall, the results showed higher total average mass concentration indoors ($7.9 \mu\text{g}/\text{m}^3$) compared to outdoors ($5.1 \mu\text{g}/\text{m}^3$).

The measured outdoor values were typical of urban sites influenced by traffic [45]. Additionally, some local sources, such as emissions from fireplaces which were used by neighbours and from adjacent fast food restaurants, have contributed to the outdoor loadings.

Table 4.2. PMC summary statistics of particle components by species over one-week period (calculated on the basis of AMS data)

	Total	Organic matter	NO ₃	SO ₄	NH ₄	Chl	Total	Organic matter	NO ₃	SO ₄	NH ₄	Chl
	Indoors (µg/m ³)						Outdoors (µg/m ³)					
Average	7,9	7,2	0,2	0,4	0,1	0,0	5,1	1,8	1,6	ktu	0,7	0,1
STD ²	28,0	27,5	0,4	0,4	0,2	0,1	3,9	1,5	1,6	0,8	0,6	0,1
I/O ratio	5,9	3,9	0,1	0,5	0,2	0,2						
Median	1,9	1,4	0,1	0,3	0,1	0,0	4,1	1,5	1,1	0,5	0,5	0,1
Maximum	1169,8	1136,7	27,4	4,4	17,4	11,5	70,6	62,4	6,2	5,3	2,5	0,6
Minimum	0,4	0,2	<0,1	<0,1	<0,1	<0,1	0,5	0,3	<0,1	<0,1	<0,1	<0,1
	Occupancy period (µg/m ³)											
	Indoors (µg/m ³)						Outdoors (µg/m ³)					
Average	6,9	6,1	0,2	0,4	0,2	0,0	5,0	1,8	1,6	0,8	0,7	0,1
STD ²	25,5	25,1	0,4	0,4	0,3	0,1	3,8	1,5	1,5	0,8	0,6	0,1
I/O ratio	1,4	3,4	0,2	0,5	0,2	0,3						
Median	2,0	1,3	0,1	0,3	0,1	0,0	4,0	1,5	1,1	0,5	0,5	0,1
Maximum	1169,8	1136,7	27,4	4,4	17,4	11,5	70,6	62,4	6,2	5,3	2,5	0,6
Minimum	0,4	0,2	<0,1	<0,1	<0,1	<0,1	0,5	0,3	<0,1	<0,1	<0,1	<0,1
	Non-occupancy period (µg/m ³)											
	Indoors (µg/m ³)						Outdoors (µg/m ³)					
Average	6,5	5,7	0,2	0,5	0,1	0,0	7,3	2,9	2,4	0,9	0,9	0,1
STD ²	5,6	6,0	0,1	0,4	0,1	0,0	4,3	1,1	1,9	0,8	0,7	0,1
I/O ratio	0,9	2,0	0,1	0,5	0,2	0,1						
Median	3,4	1,9	0,1	0,7	0,2	0,0	7,8	3,1	2,0	1,1	0,9	0,1
Maximum	25,3	25,0	0,3	1,1	0,4	25,3	14,7	4,9	5,3	2,8	2,2	0,2
Minimum	2,2	1,4	<0,1	<0,1	<0,1	<0,1	1,7	0,9	0,5	0,1	0,1	0,1

Organic matter was the dominant species indoors, accounting for most of the total mass 88 %, (Figure 4.4) with contribution from cooking, candle burning during occupancy period. The indoor mass concentration of organic matter was 3.4 times higher than outside one resulting in high I/O ratio (Table 4.2). During non-occupancy period I/O ratio have been lower for organics 1.9, due to the absence of indoor active sources, reflecting outdoor penetration, but as it was noted above, it was influenced by the concentration from previous indoor activities.

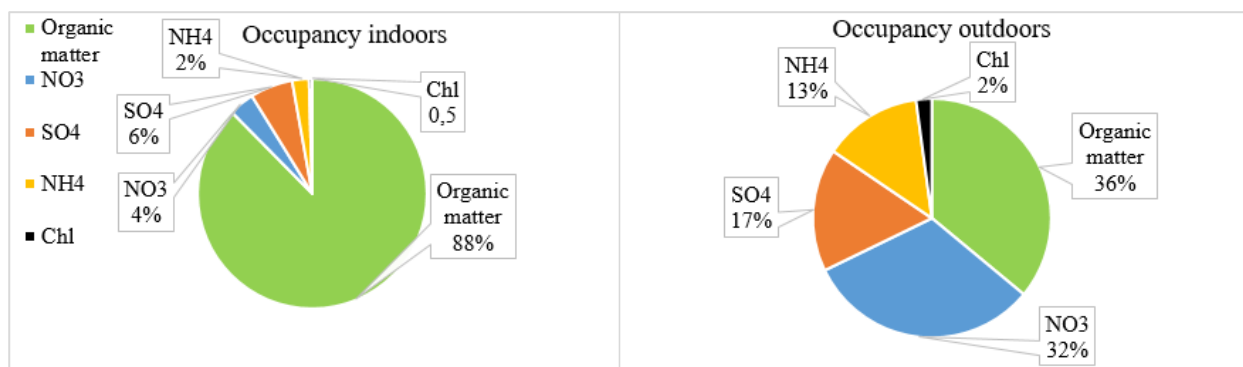


Fig. 4.4. Particle mass fractions (in percentage) by chemical species during occupancy period inside and outside of the residence

I/O ratios for nitrate accounted to 0.2, for sulphate 0.5, for ammonium 0.2, and for chloride 0.3 during occupancy time. The nitrate I/O ratio was the lowest because of its sensitivity to changes in temperature and relative humidity especially during winter period, i.e. evaporation upon moving

from a colder outdoor to a warmer indoor environment, Figure 4.4 [21; 19]. The weather conditions varied during this period with T outdoors from -1.5 to 7.9 °C and RH from 58 to 100 % and the mean (\pm standard deviation, s.d.) T_{out} was 4.3 (± 1.8) °C and RH_{out} was 86.5 (± 6) %. Indoors, T_{in} ranged from 21.5 to 26.1 °C and RH_{in} from 31.1 to 71 %, and the mean T_{in} was 24 (± 0.4) °C and RH_{in} was 41.4 (± 3.7) %.

Chloride mass loadings indoors originated mostly from outdoor environment (Figure 4.4). However, burning of certain type of the candle may be responsible for the presence of nitrate and chloride inside of the apartment (Figure 4.5B), reaching the maximum value of 27.3 $\mu\text{g m}^{-3}$ for nitrate and 11.5 $\mu\text{g m}^{-3}$ for chloride (Table 4.2). The alkali nitrate salts and zinc chloride were possibly added to the candle wick as the flame retardants, resulting in nitrate and chloride emissions [54; 44]. Alkali nitrate are known to be of water-soluble nature and if the particle of such origin end up in humid human respiratory tract it would dissolve there, bringing undesirable health effects. And possibly, nitrate having been emitted from candle could be another precursor to the formation of secondary organic species [54]. The nitrate radicals, as other type of oxidant, may react with volatile organic compounds (from indoor sources) and form secondary organic particles. But to prove it, the quantification of the nitrate concentrations is needed.

The nitrate and chloride I/O ratios within the non-occupancy period were lower, due to absence of indoor sources, showing outdoor penetration. Sulphate I/O ratio within indoor occupancy indicated that a 0.5 (or 50 %) of the outdoor origin particles has penetrated indoors, assuming the absence of indoor sulphate sources. I/O ratio during non-occupancy time were slightly higher compared to occupancy due to outdoor variations of sulphate concentrations.

Figure 4.5 A, B illustrates particle mass concentrations of organic, sulphate, nitrate, ammonium and chloride species within occupancy and non-occupancy periods inside and outside of the residence. During non-occupancy periods, low organic particle loadings can be observed (few $\mu\text{g m}^{-3}$) due to the absence of indoor sources, reflecting the result of penetration of organic particles from outdoors or residual concentration from previous indoor events. Sulphate, chloride, ammonium and nitrate mass concentrations were also low indoors as the result of outdoor penetration of sulphate and chloride chemical species, and the result of the phase change of ammonium nitrate due to the temperature and relative humidity differences upon outdoor-to-indoor transport. While when indoor sources were in operation, such as cooking – frying, deep frying, baking, candle burning, mass concentrations were increasing rapidly. Particle chemical composition dominated by organic matter during cooking, and candle burning showed high organic emissions (reaching its maximum concentration of 1136.7 $\mu\text{g m}^{-3}$), nitrate and chloride emissions.

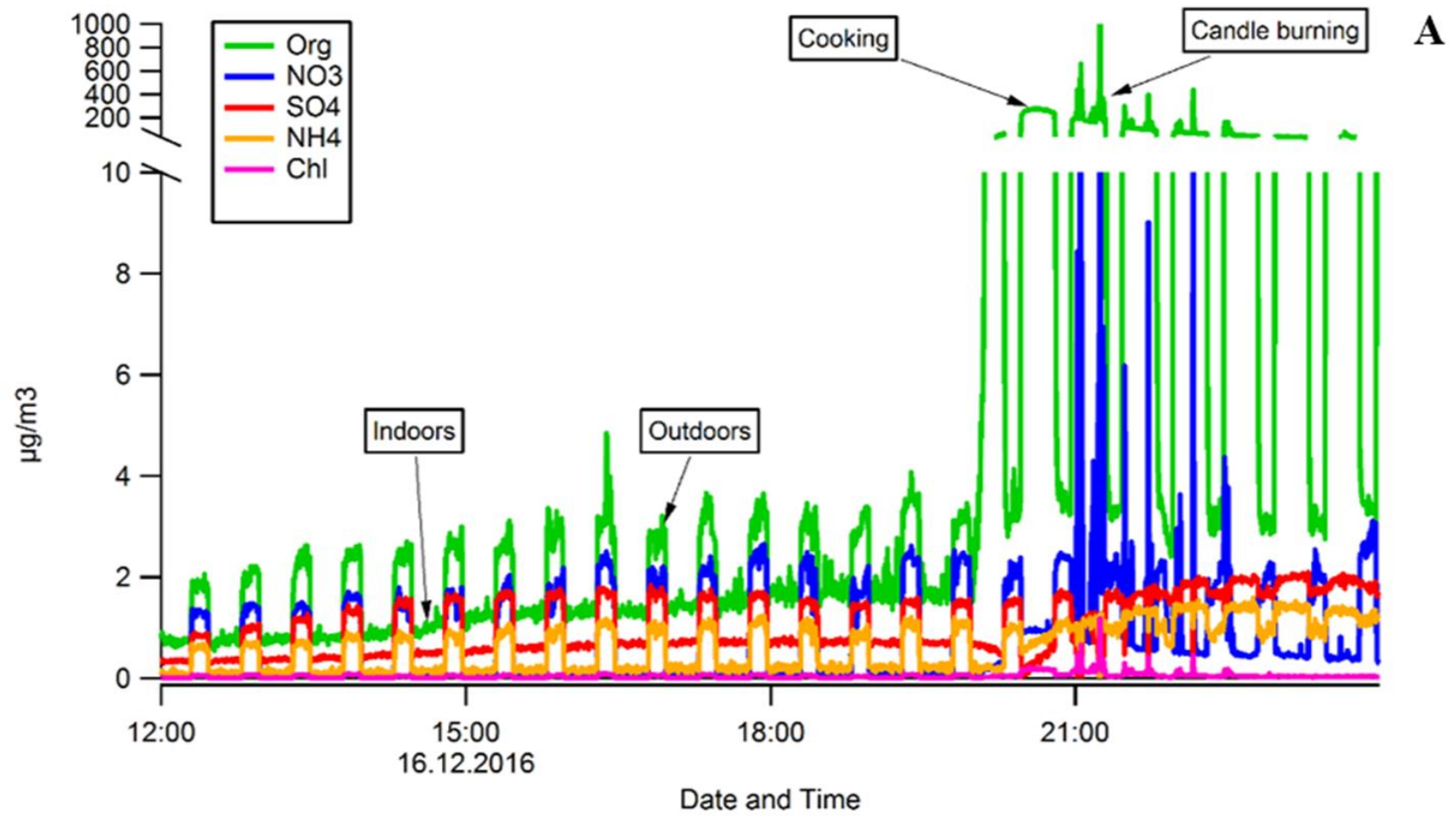


Fig. 4.5 A. Particle mass concentration during non-occupancy (12:00-18:00) and occupancy (18:00-24:00) periods inside and outside of the residence (on the basis of AMS data)

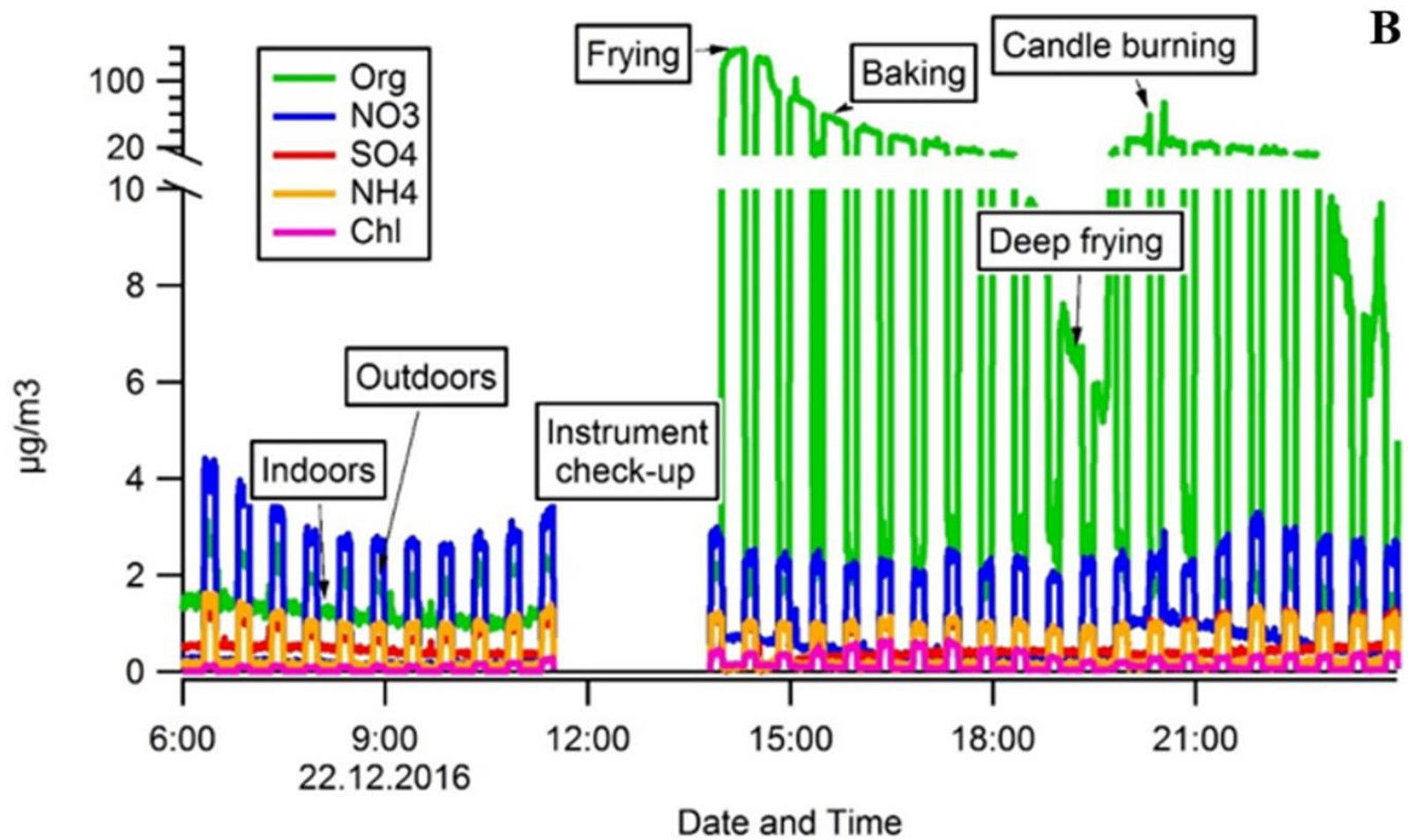


Fig. 4.5 B. Particle mass concentration during non-occupancy (6:00-12:00) and occupancy (14:15-24:00) periods inside and outside of the residence (on the basis of AMS data)

5. DISCUSSIONS AND CONCLUSIONS

The setup was successfully designed and the detailed time and size resolved particle measurements were performed inside and outside the residence in Malmo city, Sweden.

In general, particle loadings tended to be higher inside than outside of the studied apartment, mostly having been affected by the presence of active indoor sources during occupancy period (90 % out of the total monitoring period), low air exchange rate during winter season in the apartment, and variations of outdoor particle concentration. The main contributors to the particle loadings indoors were various types of cooking (frying, deep frying, baking, toasting, breakfast preparation), candle burning, ironing of the clothes. Some particles penetrated from outdoor air, forming a background level indoors, but due to considerable effect of emitted particles from indoor activities, these had the major effect. Outdoor loadings were influenced by traffic and some local sources, such as emissions from fireplaces (were used by neighbours) and from adjacent fast food restaurants. The continuous operation of indoor emission sources within occupancy period has resulted in the following PNC values, in particles cm^{-3} : average 6100, STD 23800, median 1200, max 234400, min 200. The respective to indoor occupancy outdoor values were significantly lower than inside of the apartment (in particles cm^{-3} : average 2040, STD 1208, median 1875, max 14270, min 326).

Non-occupancy period indoors showed lower number particle concentrations, in particles cm^{-3} : average 2800, STD 1800, median 2400, max 9600, min 1200, in comparison to occupancy time. The period without operation of indoor emission sources reflected outdoor penetration, however, not all the particles will penetrate indoors due to dependency in particle size. Additionally, non-occupancy period was affected by the concentration generated previously from cooking activities and particle loadings were overestimated during this time.

Indoor-to-outdoor ratio (I/O) on the basis of particle number concentration turned to be higher during occupancy time - 3.0, while during non-occupancy, with no indoor sources and no human activity indoor-to-outdoor ratio was equal to 0.9.

Particle number size distributions in the size range of 13–615 nm, inside of the apartment have exceeded the respective outdoor values during all periods of monitoring. The total monitoring and occupancy periods indoors had identical number size distributions since most of the time the occupants were present in the residence (90 % out of the total time). Occupancy period, accounting all indoor activities which were constantly occurring within analysed period and contribution of outdoor particles origin, showed to be distributed in ultra-fine particle size range (84% of total PNC), with the mode about 40 nm. The majority of the indoor sources contributed to the indoor

loadings during occupancy period also tended to emit large number of particles smaller than 100 nm. UFP have higher deposition rates in the lower respiratory tract, compared with larger particles (Hinds, 1999). During the operation of individual indoor sources (3.5 minutes SMPS scan), number size distribution during cooking resulted in PNC of 72 900 particles cm^{-3} with the mode around 85 nm, for baking 27 500 particles cm^{-3} mode around 95 nm, when making toast 19 200 particles cm^{-3} 35 nm, for ironing 135 000 particles cm^{-3} mode around 60 nm, and for candle burning 371 200 particles cm^{-3} and mode 35 nm and second mode around 130 nm (sooting burning mode).

Average number size distribution for non-occupancy period indoors reflected outdoor penetration, but due to influence of concentration from previous cooking activities size distribution shifted to larger particle sizes (mode at about 100 nm), indicating on particle growth in time because of coagulation and condensation of other components on particle surfaces.

Particle mass concentration values showed to have higher indoor values compared to outdoor ones throughout analysed period. Chemical composition of particulate matter indoors dominated by organics (88 % of the total mass during occupancy period) and this have resulted in high I/O organic ratio (3.4) due to its contribution from cooking, candle burning (reaching the maximum concentration of 1136.7 $\mu\text{g m}^{-3}$). Besides the emissions of primary emitted organic particulate matter, secondary organic species might form. Emitted particles and gases during cooking, for instance various volatile organic compounds, in the presence of the oxidants indoors (ozone, hydroxyl or nitrate radicals, could be of outdoor or indoor origin) may form oxygenated organic compounds and those with low vapor pressure might condense from the gas phase to airborne particulate matter and form secondary organic particles. Sulphate, chloride and ammonium nitrate showed to penetrate indoors from outdoor environment. Sulphate I/O ratio was within occupancy period (0.50) was slightly lower than during non-occupancy time (0.52) due to variations of outdoor sulphate concentrations, assuming the absence of indoor sulphate sources. It was found low nitrate (0.2) and ammonium (0.2) I/O ratios as semi-volatile ammonium nitrate is sensitive to changes in temperature and relative humidity parameters, i.e. dissociation and evaporation of ammonium nitrate particles upon moving from a colder outdoor to a warmer indoor environment, such phase change was especially pronounced during winter period. However, some episodic indoor source, such as candle burning may have contributed to emissions of nitrate and chloride (the maximum values during candle burning was 27.3 $\mu\text{g m}^{-3}$ and 11.5 $\mu\text{g m}^{-3}$ for nitrate and chloride, respectively). The alkali nitrate salts and zinc chloride possibly could be added to the candle wick as the flame retardants resulting in nitrate and chloride emissions. Alkali nitrate are known to be of water-soluble nature and if the particles of such origin end up in humid human respiratory tract it would dissolve there, bringing undesirable health effects. And possibly, nitrate having been emitted from candle could be another precursor to the formation of secondary organic

species. The nitrate radicals, as other type of oxidant, may react with volatile organic compounds (from indoor sources) and form secondary organic particles. But to prove it, the quantification of the nitrate concentrations with AMS high resolution analysis is needed.

The present analysis of collected data from the state-of-the-art instruments which have allowed to perform quantitative size resolved particle number (SMPS) and mass measurements (AMS) of non-refractory submicrometer particulate matter, reflects the real-time indoor concentrations of specific components from various sources and are the indicators of indoor air quality in the studied residence. Majority of the monitored active indoor sources have largely contributed to indoor loadings in terms of particle number concentration showing to be distributed in ultrafine particle size range during occupancy period. Overall, it was observed the exceedance of the indoor concentrations over the outdoor values. Chemical composition of particulate mass dominated by the organic matter; nitrate and chloride emissions could originate from episodic indoor source. Infiltrated sulphate, chloride, nitrate and ammonium particle species from outdoor sources, formed a background level indoors. Large amounts of found submicrometer-size range particulate matter of different chemical composition indoors creates potential risk in relation to the human health. Inhaled PM 2.5 have shown to be able to deposit below the tracheobronchial region, translocation of nanoparticles may potentially occur to the lung interstitium, the brain, liver, spleen and possibly to the foetus in pregnant females [82]. However, depending on the exposure time, the actual amount of translocated particles could be considerable. The detailed characterization of organic content through mass spectrometry is needed for analysis of chemical composition and toxicity particle properties, or applying Positive Matrix Factorization source apportionment approach. This will provide more information about relative contribution of different components and origin of PM to adverse health effects.

In the present thesis, the data was partly analysed based on monthly campaign within bigger project «Airborne particles in our homes: cocktail effects, chemical composition, physical characteristics and toxicity». The whole project outcomes will help to assess exposure focused on specific components and sources and define appropriate control strategy to minimize personal exposure to particulate matter indoors, possibly, as a combination of methods of removing particles generated indoors directly at the source and by preventing outdoor penetration.

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A.1 Particle losses

When making field measurements the sampling tubes are used. However, losses of submicrometer particles due to diffusion to the walls may occur. Inappropriate treatment of these losses may significantly change particle statistics. The correction for the losses for the particle size range from 13 to 615 nm was applied to the values from Scanning Mobility Particle Sizer (SMPS) by means of penetration efficiency. i.e. fraction of the particles which are able to pass through the sampling line. For instance, for the 13 nm diameter particles 31 % was lost in the tubes, Table A.1



Figure A1. Sampling lines

Penetration efficiency is expressed as:

$$P = 0.81905 \exp(-3.6568\mu) + 0.09753 \exp(-22.305\mu) + 0.0325 \exp(-56.961\mu) + 0.01544 \exp(-107.62\mu).$$

for $\mu > 0.02$

$$P = 1 - 2.5638\mu^{2/3} + 1.2\mu + 0.1767\mu^{4/3}.$$

for $\mu < 0.02$

where deposition parameter - μ :

$$\mu = \frac{\pi u L}{Q}$$

L is the length of the tube. m;

Q is the flow rate. m^3/s ; and

u - dimensionless deposition coefficient:

$$u = \frac{1.38^{23} T \eta C_c}{3 \pi \eta p_d}$$

T – temperature, K;

C_c - Cunningham slip correction factor;

η -air viscosity, Pa*s;

p_d – particle diameter, m.

Cunningham slip correction factor is applied to assume that particle moves still in continuum regime as function of particle diameter - as the particle gets smaller and closer to the gas molecule it undergo to gas collisions. To account that the small particle is moving in continuum air flow it is convenient to apply slip correction factor

$$C_c = 1 + \text{Kn}(1.257 + (0.400e^{-1.1/\text{Kn}}))$$

Kn is Knudsen number, which is needed to be accounted as it defines the extent of the contact between gas molecule and a particle:

$$\text{Kn} = \frac{2\lambda}{d_p}$$

Re number is dimensionless number that characterises the flow around a particle and is the relation between inertial forces that push away the surrounding gas (air) as the particle passes, and frictional forces between air and particle.

$$Re = \frac{F_i}{F_f} = \frac{p_a V d}{\eta}$$

p_a - air density, kg m⁻³;

η -air viscosity, Pa*s;

Table A.1 Penetration efficiency of particles through sampling lines, applied to SMPS data

Particle diameter, nm	Fraction penetrating	Particle diameter, nm	Fraction penetrating	Particle diameter, nm	Fraction penetrating	Particle diameter, nm	Fraction penetrating
13.1	0.691	34.6	0.899	91.4	0.968	241.4	0.988
13.6	0.703	35.9	0.903	94.7	0.969	250.3	0.989
14.1	0.715	37.2	0.907	98.2	0.970	259.5	0.989
14.6	0.725	38.5	0.911	101.8	0.971	269	0.989
15.1	0.736	40	0.915	105.5	0.972	278.8	0.990
15.7	0.747	41.4	0.918	109.4	0.973	289	0.990
16.3	0.757	42.9	0.922	113.4	0.975	299.6	0.990
16.8	0.765	44.5	0.925	117.6	0.976	310.6	0.991
17.5	0.776	46.1	0.928	121.9	0.976	322	0.991
18.1	0.784	47.8	0.931	126.3	0.977	333.8	0.991
18.8	0.793	49.6	0.934	131	0.978	346	0.992
19.5	0.802	51.4	0.937	135.8	0.979	358.7	0.992
20.2	0.810	53.3	0.939	140.7	0.980	371.8	0.992
20.9	0.817	55.2	0.942	145.9	0.981	385.4	0.992
21.7	0.825	57.3	0.944	151.2	0.981	399.5	0.993
22.5	0.832	59.4	0.947	156.8	0.982	414.2	0.993
23.3	0.839	61.5	0.949	162.5	0.983	429.4	0.993
24.1	0.845	63.8	0.951	168.5	0.983	445.1	0.993
25	0.852	66.1	0.953	174.7	0.984	461.4	0.993
25.9	0.858	68.5	0.955	181.1	0.984	478.3	0.994
26.9	0.864	71	0.957	187.7	0.985	495.8	0.994
27.9	0.870	73.7	0.959	194.6	0.986	514	0.994
28.9	0.875	76.4	0.960	201.7	0.986	532.8	0.994
30	0.880	79.1	0.962	209.1	0.987	552.3	0.994
31.1	0.885	82	0.963	216.7	0.987	572.5	0.995
32.2	0.890	85.1	0.965	224.7	0.987	593.5	0.995
33.4	0.895	88.2	0.966	232.9	0.988	615.3	0.995

To account of lost fractions of particle chemical species for Time-of-Flight Aerosol Mass Spectrometer (ToF-AMS) more detailed calculations are needed since determined species are undergo to condensation within the stainless-steel tubes.

The collection efficiency (CE) of the AMS is another parameter which is taken into account. Some of the mass may be lost to be due to particle bounce at the vaporizer surface before volatilization. The obtained mass concentration is divided with the CE. For liquid particles CE is 1 as liquid particles do not bounce, they splash.

Table C.1. Monitoring of occupancy/non-occupancy periods (A. Wierzbicka, 2016)

	00 - 01	01 - 02	02 - 03	03 - 04	04 - 05	05 - 06	06 - 07	07 - 08	08 - 09	09 - 10	10 - 11	11 - 12	12 - 13	13 - 14	14 - 15	15 - 16	16 - 17	17 - 18	18 - 19	19 - 20	20 - 21	21 - 22	22 - 23	23 - 24
Person A																								
Person B																								
Person C																								
Person D																								
Person E																								
Person F																								
Person G																								
Example: Person X*																								
* Person X was at home from 00:00 till 07:45 and then from 17:45 till 24:00																								