KAUNAS UNIVERSITY OF TECHNOLOGY

DARIUS ČIUŽAS

AIR QUALITY AND ITS MANAGEMENT IN LOW ENERGY RESIDENTIAL BUILDINGS

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Technological sciences, Environmental Engineering (04T)

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KAUNO TECHNOLOGIJOS UNIVERSITETAS

DARIUS ČIUŽAS

ORO KOKYBĖ IR JOS VALDYMAS MAŽAI ENERGIJOS NAUDOJANČIUOSE GYVENAMUOSIUOSE PASTATUOSE

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LIST OF ABBREVIATIONS

ACR – Air change rate;

BTEX – Benzene, toluene, ethylbenzene and xylenes;

CADR – Clean air delivery rate;

CFD – Commercial computational fluid dynamics;

CO – Carbon monoxide;

CO_2 – Carbon dioxide;

EEI – Energy efficiency index;

ELPI+ – Electric low pressure impactor;

EP – Energy performance;

EPA – Environmental protection agency;

EPBD – Energy Performance of buildings directive;

ESP – Electrostatic precipitator;

EU – European Union;

FP – Fine particle (from 0.1 to 2.5 μm);

GSS – Gas sensitive semiconductor;

HCB – Hex chlorobenzene;

HEPA – High efficiency particulate air filter;

HVAC – Heating, ventilation, and air conditioning;

IAQ – Indoor air quality;

MOS – Metal oxide semiconductor;

NMHC – Non-methane hydrocarbon;

NO_2 – Nitrogen dioxide;

NO_x – Nitrogen oxides;

O_3 – Ozone;

OP – Operating power;

OPC – Optical particle counter;

PAC – Portable air cleaner;

PAH – Polycyclic aromatic hydrocarbons;

PCB – Polychlorinated biphenyl;

PCE – Tetrachloroethylene;

PID – Photoionization detector;

PM – Particulate matter;

PM10 – Particles with a diameter smaller than 10 µm;

PM2.5 – Particles with a diameter smaller than 2.5 µm;

PNC – Particle number concentration;

RH – Relative humidity;

SBS – Sick building syndrome;

SD – Standard deviation;

SMPS – Scanning mobility particle sizer;

SPMDs – Semi-permeable membrane devices;

SVOC – Semi-volatile organic compounds;

T – Temperature;

TCE – Trichloroethylene;

TVOC – Total volatile organic compounds;

UFP – Ultrafine particle (<0.1 µm);

USA – United States of America;

UV – Ultraviolet;

UVGI – Ultraviolet germicidal irradiation;

VOC – Volatile organic compounds;

VVOC – Very volatile organic compound;

WHO – World health organisation.

INTRODUCTION

Indoor air quality (IAQ) is one of the most important parameters determining the indoor microclimate and human comfort. It is known that people in developed countries spend a majority of their time indoors (about 90%) therefore IAQ is a very important factor, but difficult to control (Brasche and Bischof, 2005; Kleipeis et al., 2001). IAQ is mostly affected by air pollution sources both indoors and outdoors. Common indoor air pollution sources are smoking, candles and incense burning, household cleaning activities, cooking, furnishing, printers, building materials and other electronic devices. Directive 2010/31/EU of the European Parliament and of the Council on the energy performance of buildings determines that by 2021 all new buildings have to be zero-energy buildings. The construction of zero-energy or low-energy houses requires a building to be airtight. Insulation creates good conditions for a higher exposure to indoor pollutants and as a result sick building syndrome (SBS) may occur. Therefore, it is important to investigate and evaluate the IAQ in low-energy houses.

IAQ may be managed by several strategies, including pollution source control, dilution (ventilation) and air cleaning. Any of these strategies requires a good understanding of indoor air pollution sources in order to obtain an effective application of engineering measures for the improvement of IAQ.

Ventilation has long been considered as one of the most effective strategies for improving IAQ, but it is often costly due to heating or cooling needs; moreover, it also introduces pollutants from outdoors. Another measure for removing pollutants from indoor air is air cleaning. It is widely applied to reduce PM and selected gaseous phase pollutants (primarily of organic origin) generated by industrial and residential sources. Ventilation and air cleaning/purification have been proven as effective ways of reducing occupant exposure to indoor contaminants and improving IAQ. At the same time, the data on the combination of both of these technologies are sparse.

The integration of novel measurement methods, new knowledge on indoor pollution sources and IAQ management strategies allow another step to be taken towards the advanced management of IAQ based on real-time pollutant sensing, and the processing of these tasks via smart home environments. The knowledge of IAQ management modelling/algorithms based on real-time sensing of specific pollutants is still very limited, thus providing an opportunity for a systematic and novel approach to this issue.

Aim of the thesis

To provide scientific background for the control of IAQ via real-time characterisation, management and modelling of indoor pollutant variations.

Objectives of the dissertation

1. To investigate IAQ in the existing low energy residential settings;
2. To characterise the dynamics of various common active indoor pollution sources via experimental modelling techniques;
3. To investigate the effectiveness of IAQ management via coupled ventilation and air cleaning;
4. To establish a control algorithm for the management IAQ based on real-time measurement data.

Scientific novelty

1. The indoor environmental quality was investigated in 11 newly built low energy residential buildings in Lithuania;
2. Unique data were obtained on the characterisation of dynamic patterns of indoor PM and VOC during various pollution episodes for real-time IAQ management;
3. A combination of ventilation and air cleaning regimes were investigated for the removal of aerosol particles and volatile organic compounds (VOCs);
4. A model has been developed for the management of IAQ based on real-time sensing of PM (nano and micro particles) and VOCs.

Structure and outline of the dissertation

This dissertation is divided into the following parts: introduction, literature review, measurements and methodology, results and discussion, conclusions, recommendations, list of 132 references, list of publications on the dissertation topic and list of annexes. The literature survey and results of the research are presented in 118 pages, including 11 tables and 21 figures.

Publication of the research results

The results of this research are presented in 3 publications, corresponding to the list of the Institute of Thomson Reuters Web of Science database, and reported at seven international conferences.

Practical value of the work

1. The data obtained from the measurements in low energy residential buildings provide a basis for the building and HVAC professionals with respect to the

insights on selecting building materials and ventilation regimes for assuring healthy IAQ.

2. The database on IAQ variation resulting from active pollution sources may serve as a basis for the further development of pollution control algorithms within buildings.

3. The created grey model for the management of IAQ in buildings based on real-time pollutant concentration measurements may be implemented in building management systems (e.g., smart home systems).

4. The results presented in the dissertation have been collected as a part of the project IAQSmart (Air quality management in Low Energy Homes), financed by the European Social Fund under the Global Grant Scheme.

Author's contribution

In the chapter "3.1. IAQ in low energy buildings in Lithuania" participation in field studies and laboratory sample processing.

In the chapter "3.2. Characterisation of active indoor pollution sources" conducting laboratory experiments, data analysis and manuscript preparation.

In the chapter "3.3. IAQ management by combined ventilation and air cleaning" conducting laboratory experiments, data analysis and manuscript preparation.

In the chapter "3.4. A grey model to IAQ management in rooms based on real-time sensing of particles and VOC" conducting laboratory experiments, data analysis and interpretation.

1. LITERATURE REVIEW

1.1. Impact of pollution sources on indoor air quality

IAQ is one of the most important parameters determining the indoor microclimate and human comfort. Because of the lifestyle induced by modern society, people tend to spend most of their time in various indoor environments such as home, workplace, or other microenvironments (e.g. transport) (Klepeis et al., 2001). On average, people spend over 90% of their time indoors, that percentage is particularly high for some specific groups, such as new-borns, the elderly, disabled or sick people. Those at the extremes of age or in poor health are likely to spend considerably more time at home than others; they may be particularly affected by changes to the indoor environment.

It is now well established that indoor air pollution contributes significantly to the global burden of disease of the population. For a majority of indoor air contaminants, particularly in the presence of common indoor sources, however, indoor concentrations usually exceed outdoor concentrations, for some pollutants even with an I/O ratio of 10 or 20 (Fernandes et al., 2008). These contaminants include a wide range of organic and inorganic substances in gaseous and particle phases (Lewis and Gordon, 1996). Exposure to these pollutants, especially fine (from 0.1 to 2.5 µm, FP) and ultrafine (<0.1 µm, UFP) airborne particles, has been identified as an important factor affecting human health (Alvin et al., 2000; Schwartz et al., 1996; Seaton et al., 1995; Jones, 1999; Dockery, 2009 and Weichenthal et al., 2007).

Air pollutants in buildings are linked to indoor factors (structure, fabrics, coating, furnishing, ventilation system, etc.), the specific activities of the occupants or to other sources found outdoors. Major sources of indoor air contaminants are:

- Ambient air pollution (due to traffic, urban and industrial activities) comes into the building through the ventilation system or by infiltration (building envelop permeability) (He et al., 2005; Hussein et al., 2005; Liu and Nazaroff, 2001 and Morawska et al., 2008);
- Building materials and furnishings (wall and floor coverings, paints, insulation materials, etc.) (Afshari et al., 2004; Kim et al., 2007 and Kelly et al., 1999);
- Processes that occur within buildings (combustion, heating, ventilation and air conditioning systems, etc.) (Gustafson et al., 2007; Wargocki et al., 2004 and Weschler, 2006);
- Occupants themselves and their activities (tobacco smoking, use of cleaning products, plant and pet treatments, cooking, paper processing such photocopying, etc.) (Afshari et al., 2005; Géhin et al., 2008; Glytsos et al., 2010; Torkmahalleh et al., 2012; Bounanno et al., 2009; Fine et al., 1999; Weschler and Nazaroff, 2008; Morawska and Zhang, 2002; Pagels et al., 2008; Isaxon et al., 2013; Stabile et al., 2012; Braver et al., 2000; Jones, 1999; Hussein et al., 2006 and Wu et al., 2012);
- Domestic animals (Raja et al., 2010 and Wright and Phipatanakul, 2014);

- Water and soil (air pollutants coming through the water supply, radon and contaminated soils) (Lamonaca et al., 2014 and Lopez et al., 2008).

These sources are commonly classified into categories such as food preparation, furniture, building materials, heating, combustion, and humans themselves (Afshari et al., 2005; Wallace, 2006; He et al., 2004 and Zai et al., 2006). They may also be classified based on their temporal variation, i.e. the rate of emission over time. Sources having rapid temporal variation may be referred as active sources (Hussein et al., 2006), such as cooking, smoking, indoor combustion, heating, use of personal care products, and application of household chemicals. The emission from building materials have a relatively slow temporal profile, thus may be referred as passive sources (Kelly et al., 1999 and Jones, 1999).

The various classifications of different indoor air pollution sources have been analysed. After this analysis, it was decided to distribute IAQ affecting factors into two groups – active and passive source. All classifications of the pollution sources are presented in **Figure 1.1.** The major pollutants formed in each process were also presented in this graph.

Indoor air pollution sources

Active

Passive

- Human generated pollution (CO_2, VOCs, microorganisms)
- Ventilation (outdoor air pollutants) (PM, NO_2, CO, VOCs, SO_2)
- Cooking (PM, NO_2, PAHs, VOCs)
- Cigarette smoking (PM, VOCs, CO, CO_2)
- Furniture polishing (VOCs)
- Car exhaust/petrol vapour (CO, NO_2, VOCs)
- Space heating (fireplaces, gas heaters, etc.) (NO_2, PM, CO, CO_2)
- Other thermal sources (candles, incense) (CO_2, PM, Pb)
- Animals (PM, allergens, microorganisms, VOCs)
- Appliances (VOCs, formaldehyde)
- Air freshener, cleaning tools and accessories toilet (VOCs, formaldehyde)

- Carpet/Carpet flooring (VOC, formaldehyde)
- Furniture made of wood particle board (VOC, formaldehyde)
- Wood dust flat floor (VOC, formaldehyde)
- Vinyl adhesive (formaldehyde)
- Walls painted with emulsion (VOC, formaldehyde)
- Cavity wall insulation (VOC, formaldehyde)
- Printing ink (VOC)

Figure 1.1. Indoor air pollution sources affecting IAQ.

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1.2.   Active sources of indoor air pollution

Active sources of indoor air pollution affect IAQ the most, because they release a lot of gaseous compounds and PM in a relatively short period of time.

Cooking related sources

Food preparation activities have been reported as substantial emitters of UFP and FP. Different working groups were able to detect very high pollution peaks during cooking with electric stoves and in particular gas stoves at 1×10^5 to 5.6×10^5 p/cm^3 (Afshari et al., 2005; Géhin et al., 2008; Glytsos et al., 2010; Torkmahalleh et al., 2012; Bounanno et al., 2009; He et al., 2004 and Hussein et al., 2006). The large concentration range is attributable to the different cooking activities (e.g. baking, roasting, frying and toasting), the use of energy, the respective cooking goods, the ventilation conditions and the room geometry. Dennekamp et al. (2001) describe particle number concentrations of up to 1.1×10^5 or 1.5×10^5 p/cm³ when using four electric or gas rings. Peak values of up to 5.9×10^5 UFP/cm³ were reached during the frying of bacon on a gas stove. After a short period of time the particles increased in the indoor air with a displacement towards larger diameters (Abt et al., 2000; Dennekamp et al., 2001 and Hussein et al., 2006). Extremely high levels of pollution are to be expected when cooking on open fireplaces as, for instance, in third world countries (e.g. Naeher et al., 2000). See and Balasubramanian (2006 a, b) performed controlled experiments in a domestic kitchen using five different gas-cooking methods. Their results showed that deep-frying produced the highest concentration of particles, up to 6.0×10^5 p/cm³, and the mode diameter was approximately 20 nm. Yeung and To (2008) examined the size distribution of aerosols emitted from commercial cooking activities. Particle number concentration measurements revealed that the particle size distribution followed a lognormal distribution and that the aerosol mode diameter increased as cooking temperature increased.

Burning sources

The major categories of products resulting from combustion can be listed as carbon monoxide (CO), nitrogen oxides (NOx), particulate material, and polynuclear aromatic hydrocarbons (PAH).

Combustion sources such as candles, incense and cigarette burning have been identified as sources of VOCs and UFP. Candles and incense burning have been identified as emitting particles in the diameter range of 0.03 – 3 µm (Fine et al., 1999; Weschler and Nazaroff, 2008; Morawska and Zhang, 2002; Pagels et al., 2009; Isaxon et al., 2013 and Stabile et al., 2012). Emissions from tobacco smoking have attracted considerable attention as both a source of particles and VOCs (Brauer et al., 2000; Jones, 1999; Afshari et al., 2005; Glytsos et al., 2010; Hussein et al., 2006 and Wu et al., 2012). The particle number increases considerably during cigarette smoking, partly to values up to 2.1×10^5 p/cm³ (He et al., 2004; Afshari et al., 2005 and Hussein et al., 2006). A lighted cigarette gives off approximately 4,700 chemical compounds (benzene, acetone, formaldehyde, phenols, ammonia, aromatic hydrocarbons, toluene and etc.). Benzene, a known human carcinogen, is generally

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emitted from synthetic fibres, plastics and some cleaning solutions; however, the most important exposure is from cigarettes. Wallace reported that exhaled benzene was 10 times higher for smoker than non-smoker households (Wallace, 2000). When burning candles or oil lamps in indoor environments, an increase in UFP was likewise observed (Fine et al., 1999; Hussein et al., 2006 and Wallace and Ott, 2011). This involved significantly higher concentrations when extinguishing candles compared to the burning itself (Hussein et al., 2006). During the burning of incense sticks it is also possible to detect high particle contents, in particular in the range from 0.06 to 2.5 μm, in indoor air (Chao et al., 1998 and Jetter et al., 2002).

Household sources

During cleaning and in particular vacuum cleaning, an increase in coarse particles, and in particular the particle mass, is observed in indoor air (Abt et al., 2000). Long et al. (2000) investigated the influence of the use of commercial cleaning agents, based on pine oil, on the exposure in a living room. During the activities the particle number concentrations rose from an initial 2×10^3 p/cm³ to a maximum of 1.9×10^5 p/cm³ and the PM2.5 contents increased from 5 to 38 μg/m³. This phenomenon was explained by referring to the new particle formation or particle growth through oxidative processes in the indoor environment. Other working groups, too, were able to detect the presence of ozone in test chambers, and the simultaneous application of terpene-containing cleaning agents caused a significant increase in particle number concentrations and the particle mass (Singer et al., 2006). Air-freshener sprays also produce substantial amounts of particle mater (Afshari et al., 2005), due to the reaction of VOCs with ozone that lead to nucleation of UFP (Nazaroff and Weschler, 2004). Vacuum cleaners operating with or without dust bags have also been shown to produce particle mater in a size range above 0.3 μm (Lioy et al., 1999).

He et al. (2004) reported a single event peak number concentration of 9.5×10^3 p/cm³ during experiments in residential houses, while Hussein et al. (2006) did not observe any increase in particle concentration during the use of a hair dryer. Glytsos et al. (2010) reported that hair spray experiments showed that mainly particles larger than 1 mm were emitted. The average increase in the particle number concentration was 1.5 times the background concentration, while the average PM2.5 increase was 11.6 times the background.

1.3. Health effects of indoor air pollution

1.3.1. Indoor air pollution and health

IAQ refers to the air quality within and around buildings and structures, especially as it relates to the health and comfort of the building occupants. Indoor air contaminants can originate within a building or be drawn in from outdoors. Understanding and controlling common pollutants indoors can help reduce the risk of indoor health concerns. Symptoms of poor IAQ are very broad and depend on the contaminant. They can easily be mistaken for symptoms of other illnesses such as

allergies, stress, colds and influenza. Poor IAQ can lead to a number of physical symptoms and complaints. The most common include:

- Coughing;
- Sneezing;
- Watery eyes;
- Fatigue;
- Dizziness;
- Headaches;
- Upper respiratory congestion.

The effects of indoor air pollutants range from short-term effects; eye and throat irritation; to long-term effects; respiratory disease and cancer. Exposure to high levels of some pollutants, such as carbon monoxide, can even result in immediate death. Some indoor pollutants can magnify the effects of other indoor pollutants (USEPA, 2013).

Short-term health effects may show up shortly after a single exposure or repeated exposures to a pollutant. These include irritation of the eyes, nose, and throat, headaches, dizziness, and fatigue. Such immediate effects are usually short-term and treatable. Sometimes the treatment is simply eliminating the person's exposure to the source of the pollution, if it can be identified. Soon after exposure to some indoor air pollutants, symptoms of some diseases such as asthma may show up, be aggravated or worsened. Certain immediate effects are similar to those from colds or other viral diseases, so it is often difficult to determine if the symptoms are a result of exposure to indoor air pollution. For this reason, it is important to pay attention to the time and place symptoms occur. If the symptoms fade or go away when a person is away from the area, for example, an effort should be made to identify the indoor air sources that may be possible causes. Some effects may be made worse by an inadequate supply of outdoor air coming indoors or from the heating, cooling or humidity conditions prevalent indoors (Berglund et al., 1992).

Long-term health effects may show up either years after exposure has occurred or only after long or repeated periods of exposure. These effects, which include some respiratory diseases, heart disease and cancer, can be severely debilitating or fatal (USEPA, 2013).

The composition of chemicals known collectively as indoor air pollutants is a complex array of constituents made up of gases, vapours and particles. Determining the health effects related to these pollutants collectively, individually, or in certain combinations requires extensive information about the exposure of an individual to this mixture. A body of literature is developing that provides information in varying degrees on the major indoor pollutants affecting human health. These pollutants can be divided into particles (solids and liquid droplets) and vapours and gases (Burroughs and Hansen, 2008).

1.3.2. Particulate matter and health

Particulate air pollution is a mixture of solid, liquid or solid and liquid particles suspended in the air. These suspended particles vary in size, composition

and origin. Indoor particles consist of ambient particles that have infiltrated indoors, particles emitted indoors and particles formed indoors through reactions of gas-phase precursors emitted both indoors and outdoors (Meng et al., 2005; Uhde and Salthammer, 2007 and Morowska and Salthammer, 2003). The sizes of particles encountered in indoor environments range from molecular clusters of a few nm in diameter to larger than 10 µm. Key attributes of particles: sources, composition, dynamic, dynamic behaviour, fates and effects, are all strongly related to particle size (Nazaroff, 2004). According to the size spectrum particles can be divided into two modes: fine (particle smaller than 2.5 µm, frequently referred as PM$_{2.5}$, which stands for PM with a diameter smaller than 2.5 µm), and coarse (particles with a diameter greater than 2.5 µm). Often an ultrafine mode is also distinguished, comprised of particles with diameter smaller than 100 nm (**Figure 1.2**).
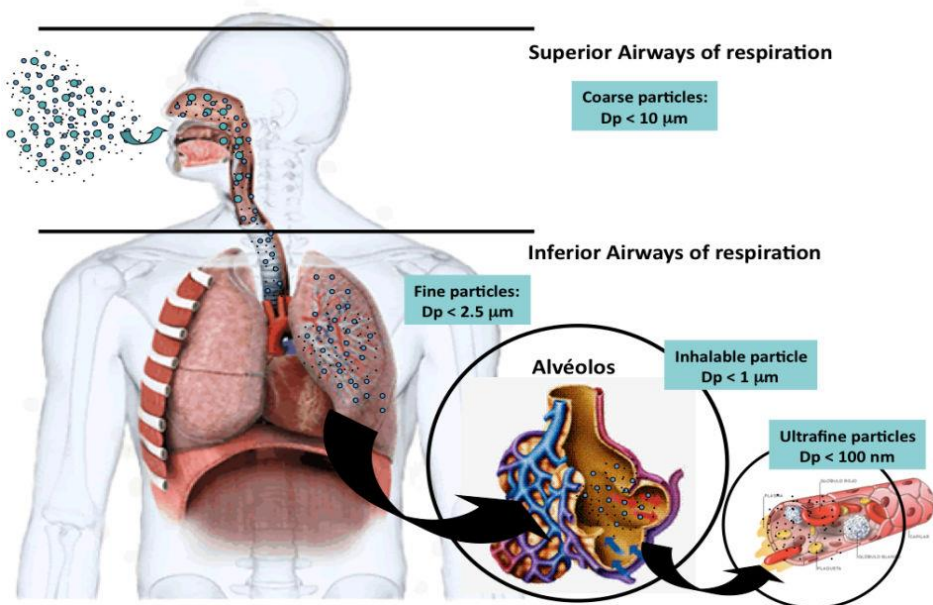
Figure 1.2. Represents the areas where PM is deposited in the body (Guarieiro and Guarieiro, 2013).

The health hazard caused by inhaled particles depends on many factors, such as particles size, chemical composition, number, surface area, morphology, hygroscopicity and the site on which they are deposited within the respiratory system. The most important of the are: impaction, sedimentation and thermal/turbulent diffusion. Particles that contact the airway walls are deposited there and are not exhaled.

Particle size is directly related to its potential for causing health problems. Small particles less than 2.5 µm in diameter can be inhaled deeper into the lungs. Scientific studies have linked exposure to high concentrations of some types of particle mater with a variety of problems, including:

- Death from respiratory and cardiovascular causes, including strokes (Dominici et al., 2002; Tsai et al., 2003 and Wellenius et al., 2005);
- Increased mortality in infants and young children (Pope and Dockery, 2006);
- Increased numbers of heart attacks, especially among the elderly and in people with heart conditions (Zanobetti and Schwartz, 2005);
- Inflammation of lung tissue in young, healthy adults (Ghio et al., 2000);
- Increased hospitalisation for cardiovascular disease, including strokes and congestive heart failure (Metzger et al., 2004 and Wellenius et al., 2006);
- Increased emergency room visits for patients suffering from acute respiratory ailments (Van Den Eeden et al., 2002);
- Increased hospitalisation for asthma among children (Lin et al., 2002 and Norris et al., 1999);
- Increased severity of asthma attacks in children (Slaughter et al., 2003).

People with heart or lung diseases, children and older adults are the most likely to be affected by particle pollution exposure.

1.3.3. VOCs and health

Organic compounds that exist as a gas, or can easily off-gas under normal room temperature and relative humidity, are considered volatile. A range of VOCs are always present in indoor air. While most people can smell high levels of some VOCs, others have no odour. Odour does not indicate the level of risk from inhalation of this group of chemicals. There are thousands of different VOCs produced and used in today's homes. There are estimated to be 50 to 300 different compounds that could be classed as VOCs in a typical domestic indoor environment. The list of potential sources is lengthy and growing. Some common examples include:
- Acetone;
- Benzene;
- Ethylene glycol;
- Formaldehyde;
- Methylene chloride;
- Perchloroethylene;
- Toluene;
- Xylene;
- 1, 3-butadiene.

The United States Environmental Protection Agency (USEPA) has found concentrations of VOCs in indoor air to be 2 to 5 times greater than in outdoor air and sometimes far higher. During certain activities indoor levels of VOCs may reach 1,000 times that of the outside air. Studies have shown that individual VOC emissions by themselves are not that high in an indoor environment, but the indoor total VOC (TVOC) concentrations can be up to five times higher than the VOC levels outdoors (Jones, 1999). New buildings especially, contribute to the highest level of VOC off-gassing in an indoor environment because of the abundant new materials generating VOC particles at the same time in such a short period (Wang et

al., 2007). VOC concentrations in an indoor environment during winter are three to four times higher than the VOC concentrations during the summer. High indoor VOC levels are attributed to the low rates of air exchange between the indoor and outdoor environment as a result of tight-shut windows and the increasing use of humidifiers (Schlink et al., 2004). Many products we have in the home release or "off-gas" VOCs. Some examples of sources of VOCs are:

- Buildings material (carpets and adhesives; composite wood products; paints; sealing caulking; solvents, upholstery fabrics; varnishes and vinyl floors);
- Home and personal care products (air fresheners; air cleaners that produce ozone; cleaning and disinfecting chemicals; cosmetics; fuel oil, gasoline; moth balls and vehicle exhaust from running a car in an attached garage);
- Human behaviour (cooking; dry cleaning; hobbies; newspapers; non-electric space heaters; photocopiers; smoking; stored paints and chemicals and wood burning stoves).

The ability of VOCs to cause health effects varies greatly. As with other chemicals, the effects of VOC exposure depend on several factors including the type of VOC, the amount of VOC and the length of time a person is exposed. Each chemical has its own toxicity and potential for causing different health effects. Common symptoms of exposure to VOCs include: short-term exposure to high levels of VOCs (eye, nose and throat irritation; headaches; nausea/vomiting; dizziness; Worsening of asthma symptoms) and long-term exposure to high levels of VOCs (increased risk of: cancer; liver damage; kidney damage; central nervous system damage). Some people do not appear to have any kind of reaction to relatively "low" amounts of VOCs, while other people are fairly sensitive. Since many people spend much of their time indoors, long-term exposure to VOCs in the indoor environment can contribute to sick building syndrome (SBS) (Vermont Department of Health). Of course the building is not really sick, but it has developed a condition that can make its occupants uncomfortable, irritated, or even ill.

SBS is diagnosed when complaints and symptoms are clearly associated with building occupancy, but no causal agent can be positively identified. Many SBS problems are associated with new constructions. Symptoms include respiratory complaints, irritation and fatigue. Sensory perception of odours leads to a perception of poor air quality and consequently stress initiated behavioural responses (e.g. opening windows). Other environmental stressors such as noise, vibration, crowding, ergonomic stressors and inadequate lighting can produce symptoms similar to those caused by poor air quality. The negative effects can reduce productivity in offices and learning ability in schools.

1.4. Impact of energy efficiency on IAQ

1.4.1. Low energy buildings

Domestic and international efforts are needed in order to mitigate climate change. It has been estimated that the largest potential for energy saving and

decreasing greenhouse gas emissions is in the building sector, where some 27% of energy is used in residential buildings (Commission, 2007).

Buildings exist primarily to provide a healthy and comfortable environment in which people can work and live. The indoor environment must be maintained at a comfortable and healthy temperature, have an adequate supply of fresh air, be free from damp, draughts and pollutants and be quiet and well lit. A comfortable indoor environment has to rely on the use of energy for lighting, ventilation, heating and/or cooling. While heating has traditionally been the major cause for energy consumption in most European countries, HVAC systems for cooling have been installed in recent years at increasing rates in Southern European countries. Also, cooling is often needed in office buildings all year round because of high internal gains due to computers and artificial lighting. This suggests that natural lighting strategies have an important bearing on the balance between heating and cooling needs of a building in most latitudes.

The aim is to combine healthy, comfortable indoor environments with the least amount of energy required to achieve this, taking the rational use of resources into account.

Low-energy buildings typically use high levels of insulation, energy efficient windows, low levels of air infiltration and heat recovery ventilation to lower heating and cooling energy. They may also use passive solar building design techniques or active solar technologies. These homes may use hot water heat recycling technologies to recover heat from showers and dishwashers. Lighting and miscellaneous energy use is alleviated with fluorescent lighting and efficient appliances.

The assessment of energy efficiency follows nationally agreed methods of estimating energy consumption (EU 2012), following a comparative methodology framework the EU has established for calculating cost-optimal levels of minimum energy performance requirements for buildings and building elements. The methods in use have evolved in the past decades and they vary across European countries.

In Lithuania, implementation of the Energy Performance of Buildings Directive (EPBD) started when certification requirements for new buildings came into force on January 1, 2007. The latest requirements for new buildings in relation to EPBD recast became effective on January 9, 2013. Energy performance requirements are not obligatory for existing buildings, which are for sale or rented, but the evaluation procedure and certification requirements for existing buildings after major retrofits have been required since January 1, 2009.

Buildings are classified into nine energy performance (EP) classes, ranging from A++ (nearly zero-energy buildings) to G (energy-inefficient). The evaluation of buildings does not refer to their purpose of use, but to their technical specifications.

The meaning of the term 'low-energy house' has changed over time, but in Europe it generally refers to a house that uses around half of the German or Swiss low-energy standards referred to below for space heating, typically in the range from 30 kWh/m^{2}a to 20 kWh/m^{2}a). In Lithuania a low-energy house is represented by B,

A and A+ energy performance classes as defined by the national standard STR 2.01.09:2012 implementing the directive 2010/31/EU (the total primary energy use to 85 kW/m^{2}a).

The countries within the EU have assumed commitments to build low energy consumption buildings from 2016 to 2020 (Directive, 2010). The Directive 2010/31/EU of the European Parliament and of the Council on the energy performance of buildings determines that by 2020 all new buildings have to be zero-energy buildings (Commission, 2007).

1.4.2. IAQ in low energy buildings

The concentration of an indoor air pollutant depends not only on its indoor emission rate, but also on the rate at which it is being transported from outdoors to indoors, and the rates at which it is scavenged by indoor surfaces, consumed by indoor chemistry, and removed by ventilation or air cleaning (Weschler, 2009). Recently, the removal of pollutants via ventilation is being challenged due to increasing occurrence of energy efficient buildings due to increasing demand in energy saving. Airtightness of buildings and presumably decreasing ventilation may have a negative impact on indoor air quality, however little attention has been paid to the IAQ in low-energy buildings in comparison to conventional buildings.

Derbez et al., (2014), investigated IAQ and comfort in seven newly built energy-efficient buildings in France. The authors reported that compared to standard French buildings, the concentrations of benzene, ethylbenzene, m- and p-xylenes, PM2.5 and radon were low, whereas the CO_2 and formaldehyde levels were not significantly different. In contrast, the levels of acetaldehyde, hexaldehyde, n-decane, n-undecane, o-xylene and styrene were higher in these new homes, possibly because of the emissions from products and materials (Derbez et al., 2014). In a subsequent study Langer et al., (2015), investigated IAQ in 20 new passive buildings and 21 new conventionally built buildings in Sweden. Median concentrations in the passive and the conventional buildings were 10 and 12 µg/m^3 for NO_2, 9.7 and 11 µg/m^3 for ozone, 11 and 16 µg/m^3 for formaldehyde, and 270 and 150 µg/m^3 for TVOC, respectively (Langer et al., 2015). Significant differences in the TVOC and formaldehyde concentrations between the two groups of buildings indicated substantial sources of TVOC present in the passive buildings. Kolarik et al., (2012), investigated concentrations of formaldehyde in 20 new Danish residential buildings and reported that in two buildings formaldehyde concentrations exceeded WHO guideline values (Kolarik et al., 2012). Brown, (2000), investigated IAQ in new and established buildings in Melbourne, Australia, and showed that the airborne VOC levels were one to two orders of magnitude higher in new or renovated buildings than in established dwellings (Brown, 2002).

Urban et al., (2006), describe an investigation of the first low energy building constructed in the Czech Republic. It is a two-floored building with nine apartments served by mechanical ventilation with heat recovery. The study involved a questionnaire survey of the occupants and evaluation of the filters removed from the HVAC. Analysis of the filters did not reveal unexpected occurrences of fungi, pollen

and bacteria. The questionnaire study identified a problem with a lack of information for users about the functioning of the heating, HVAC unit, the effect of its use and the purpose of the forced ventilation. The mechanically ventilated buildings were generally perceived as unhealthier and less comfortable for the user. There were reports of smell diffusion between apartments from the kitchen, a high level of noise from the HVAC and fungi were a problem in some apartments.

Yoshimo et al., (2006), report that indoor air pollution by chemical substances is a serious problem in Japan and that such pollution is associated with SBS. They investigated 60 houses where occupants were suspected of suffering symptoms caused by indoor pollution. Concentrations of VOCs and formaldehyde as well as air exchange rates (in some) and airtightness were measured. Only 36% of the 14 homes met the 0.5 ACH required in the Building Standard Law, others being below this value. The concentration of formaldehyde, toluene, ethylbenzene, xylene, p-dichlorobenzene and TVOC was significantly higher in SHS homes. Concentrations were higher in new homes or following refurbishment, in those with high airtightness and low air exchange rate, and where there was new furniture or moth crystals were used.

1.5. Pollutant control strategies

Good IAQ may be achieved by multiple measures, among which are ventilation (dilution with less contaminated air), air cleaning, prevention (using low emitting building materials, avoiding combustion sources indoors, cautiously using personal care products etc.) (Han et al., 2014 and Zhang et al., 2011). Filtration and air cleaning can be a truly cost-effective and useful tool in IAQ prevention and mitigation.

1.5.1. Ventilation

All homes need ventilation (the exchange of indoor air with outdoor air) to reduce indoor moisture and indoor sources (e.g. human metabolism, occupant activities, consumer products, furnishing, building equipment and materials). Dilution is a preferred control when the contaminant/source is unknown, source treatment is too costly, or when the source is not localised. Ventilation control means much more than the amount of outside air brought into a facility. It includes the quality of outside air; the effectiveness with which it reaches occupants; and its efficiency in reducing contaminant levels. Contaminants such as formaldehyde, VOCs, and radon that may cause health problems can accumulate in poorly ventilated homes. Inadequate ventilation prevents unpleasant doors from being removed. Excess moisture generated within the home needs to be removed before high humidity levels lead to physical damage to the home or mould growth.

Ventilation techniques and strategies have become more sophisticated as homes have become more complex. Ventilation is characterised by ACH (h^{-1}), which shows how many times the air inside a defined space (room, house, or facility) is being replaced by outside air within one hour. The American Society of Heating, Refrigerating and Air-Conditioning Engineers, Inc. (ASHRAE) requires

that natural, or natural plus mechanical, ventilation provide a whole-house ventilation rate of 0.35 air changes per hour (ACH), but not less than 15 cubic feet per minute (cfm) per person. Typical air change rates measured in houses in US were found in a range of $0.5 - 2.2$ h^{-1} varying with room type and season (Du et al., 2012). Ventilation rates greater than 4 h^{-1} are usually avoided in residential environments (Yamamoto et al., 2010).

Natural ventilation is the process of supplying air to and removing air from an indoor space without using mechanical systems. It refers to the flow of external air to an indoor space as a result of pressure differences arising from natural forces. There are two types of natural ventilation occurring in buildings: wind driven ventilation and buoyancy-driven ventilation. Wind driven ventilation arises from the different pressures created by wind around a building or structure, and openings being formed on the perimeter which then permit a flow through the building. Buoyancy-driven ventilation occurs as a result of the directional buoyancy force that results from temperature differences between the interior and exterior.

Mechanical ventilation uses fans to drive the flow of outside air into a building. This may be accomplished by pressurisation (in the case of positively pressurised buildings), or by depressurisation (in the case of exhaust ventilation systems). Many mechanically ventilated buildings use a combination of both, with the ventilation being integrated into the HVAC system. The role of the HVAC system in delivering the required thermal conditions and meeting IAQ ventilation needs is a critical IAQ concern.

Heat recovery ventilation can reduce energy use for heating and cooling by facilitating a sensible heat exchange between exhaust air and incoming ventilation air. Energy recovery ventilation transfers moisture in addition to sensible heat. However, heat recovery can increase the fan power required for ventilation, and may increase energy use for heating and cooling for periods when ventilation would be beneficial for the control of indoor thermal comfort.

1.5.2. Air cleaning

Air cleaners employing filters are of two general types. Portable air cleaners (PAC) are self-contained devices that can be placed in a room, such as the bedroom of an allergic child, and operated there to reduce particle levels in the room. In-duct air cleaners are installed in the ductwork of a home with central forced air, and are designed to reduce particle levels throughout the house.
Air cleaning technology:
- Mechanical filtration;
- Electret filtration;
- Electrostatic precipitation;
- Sorption (activated charcoal, alumina, etc.);
- Ionisation (negative, bipolar);
- Photocatalytic oxidation;
- Ultraviolet germicidal irradiation;
- Ozone generation;

- Hybrids (combinations of the above).

Mechanical filtration

Air filtration is emerging as an important element in IAQ. The mechanical filter simply intercepts particles in the air passing through the filter. The most common type of filter is the fibrous filter. Fibrous filters consist of layers of fibres, crisscrossed in more or less random ways so as to allow air molecules to find a tortuous path through the filter, while at the same time removing particles that intercept the fibres. The thicker or more tightly weaved the filter, the more success it will have in removing particles. However, increased thickness or tightness of the weave requires more energy to move the air through the filter, so there is an economic cost in going to more efficient mechanical filters.

High efficiency particulate arrestors (HEPA) filters usually consist of extremely densely packed fiberglass. To obtain the designation HEPA, they must filter out 99.97% of particles 0.3 μm in diameter. This particle size is normally the hardest to filter, so the efficiency should be even greater than 99.97% for all other sizes. "HEPA-type" filters meet a less stringent standard of 95% efficiency for 0.3 μm particles (Wallace, 2008).

The effectiveness of media filters changes as PM builds up. If left alone, they gradually increase in collection efficiency. That, however, is not a good reason to leave them unattended. As they clog, the flow rate decreases. Resistance from clogged filters reduces air flow and system efficiency and increases energy usage.

Electret filtration

The electret filter employs an electrical charge to enhance collection of particles. For example, the fibres can be permanently charged. Since some air particles are charged, they will be attracted to charges of opposite polarity on the filter. Even for uncharged particles, the existence of a charge on a nearby fibre will induce a dipole moment (pushing like charges to the back of the particle and unlike charges to the front, as seen from the fibre) and the net result is an attraction to the fibre. Since the electric charge on the filter does not affect the pressure drop, there is no energy penalty. Electrets are widely used in automobiles to filter air coming in to the passenger compartment (Wallace, 2008).

Electrostatic precipitation

An electrostatic precipitator (ESP) has demonstrated great efficiency in removing particles in the range 0.01 μm to 5.0 μm, like dust and smoke, from a flowing gas using the force of an induced electrostatic charge minimally impeding the flow of gases through the unit. An electric charge is to actively add charge to the incoming particles by passing them through a strong electric field created by multiple wires. The particles are driven to a number of collecting plates. The collected material must, however, be regularly removed from the collector plates.

Sorption

Activated carbon is a porous material that can adsorb volatile chemicals on a molecular basis, but does not remove larger particles. The adsorption process when using activated carbon must reach equilibrium thus it may be difficult to completely remove contaminants (Zeltner et al., 2005). Activated carbon is merely a process of changing contaminants from a gaseous phase to a solid phase, when aggravated or disturbed contaminants can be regenerated in indoor air sources (Ao et al., 2004). Activated carbon can be used at room temperature and has a long history of commercial use.

Ionisation

An ioniser use charged electrical surfaces or needles to generate electrically charged air or gas ions. These ions attach to airborne particles which are then electrostatically attracted to a charged collector plate. This mechanism produces trace amounts of ozone and other oxidants as by–products. Most ionisers produce less than 0.05 ppm of ozone, an industrial safety standard.

Photocatalytic oxidation

Photocatalytic oxidation systems are able to completely oxidise and degrade organic contaminants. For example, VOCs found at low concentrations, within a few hundred ppm or less, are the most likely to be completely oxidised. Photocatalytic oxidation uses short-wave ultraviolet light (UVC), commonly used for sterilisation, to energise the catalyst (usually titanium dioxide (TiO_2) and oxidise bacteria and viruses. Photocatalytic oxidation in-duct units can be mounted to an existing forced-air HVAC system. Photocatalytic oxidation is not a filtering technology, as it does not trap or remove particles. It is sometimes coupled with other filtering technologies for air purification. UV sterilisation bulbs must be replaced about once a year; manufacturers may require periodic replacement as a condition of warranty.

Ultraviolet germicidal irradiation

Ultraviolet germicidal irradiation (UVGI) can be used to sterilise air that passes UV lamps via forced air. Air purification UVGI systems can be freestanding units with shielded UV lamps that use a fan to force air past the UV light. Other systems are installed in forced air systems so that the circulation for the premises moves micro-organisms past the lamps.

Ozone generation

Ozone generators are designed to produce ozone, and are sometimes sold as whole house air cleaners. Unlike ionisers, ozone generators are intended to produce significant amounts of ozone, a strong oxidant gas which can oxidise many other chemicals. The only safe use of ozone generators is in unoccupied rooms, utilising "shock treatment" commercial ozone generators that produce over 3,000 mg of ozone per hour. However, it is not healthy to breathe ozone gas, and one should use extreme caution when buying a room air purifier that also produces ozone.

Hybrids

There can also be hybrid technology. For example, some electrets may also have a continuous electric field applied to them, in an attempt to reduce the loss of charge over time that has been noted for electret fibres.

For removing pollutants from indoor air PAC can be used. This form of pollution control is widely applied to reduce PM and selected gaseous phase pollutants (primarily of organic origin) generated by industrial and residential sources. PAC generally contains a fan to circulate the air and uses one or more of the air cleaning devices discussed above. PAC may be moved from room to room and used when continuous and localised air cleaning is needed.

There are numerous technologies used in such devices including filtration, electrostatic precipitation, ion generation, pulsed discharge plasma combined with photo-catalyst, etc. (Chen et al., 2005; Grinshpun et al., 2005; Kim et al., 2012 and Shiue et al., 2011). PACs have been used in indoor environments to remove smoke from tobacco, wood stoves and fireplaces, VOCs from cleaning products, personal care products, and new furniture (Jung et al., 2013; Shaughnessy and Sextro, 2006 and Zhang et al., 2011). PACs are popular air cleaning devices that are used in 10-30% of American homes (Shaughnessy and Sextro, 2006). Hence, air cleaners are looked at not as the replacement for ventilation, but rather a supplement.

The comparison of the PAC is performed based on the specific standards and metrics that allow uniform evaluation. One such metric is the clean air delivery rate (CADR), which is the effective volumetric flow rate of pollutant free air delivered by the PAC. The CADR is considered as the best available metric to compare PACs because it takes into account the flow rate through the air cleaner, the volume of the test room, and the pollutant removal efficiency (Shaughnessy and Sextro, 2006). In the case of particles, the CADR was found around 40 m^3h^{-1} for an ion generator, 70 m^3h^{-1} for an electrostatic precipitator, and ranging from 100 to almost 300 m^3h^{-1} for the three filter-based air cleaners for particles with diameters above 100 nm (Mølgaard et al., 2014). At the same time, PACs are much less efficient in removing organic compounds from indoor air. For example, Kim et al., (2012), examined the gas-removal performance of 18 room air cleaners and determined CADR ranging from 6 (acetaldehyde) to 72 m^3h^{-1} (toluene), with a removal efficiency ranging from 20 to 90%, respectively. Chen et al., (2005), examined the gas-removal performance of 15 room air cleaners and determined CADR ranging from 0.3 (dichloromethane) to 129 m^3h^{-1} (1.2–dichlorobenzene), with removal efficiency ranging from 0.1 to 43%, respectively.

1.5.3. Prevention – source control

The most effective way to improve IAQ is to eliminate individual sources of pollution or to reduce their emissions. In many cases, source control is also a more cost-efficient approach to protecting IAQ than increasing ventilation because increasing ventilation can increase energy costs. Examples include not smoking indoors and removing animals, unused solvents and paint thinners from the home. Point sources of human-produced pollutants can often be controlled by localised

exhaust (such as bath and kitchen fans and duct collectors in a shop). Damp basements and other areas where standing water is prevalent can become the breeding ground for pollutants such as mould, mildew and other microbes.

Source control should be exercised during building operations, maintenance, and renovation as well as during initial design. Designers should specify building materials that are minimal sources of indoor emissions. These materials include low-emitting products and materials, which do not generate or store dust particles. In addition, the design should minimise horizontal surfaces on interior finishes and furnishings to reduce the particle levels in buildings.

The concentration of VOC within the air of a building can be controlled through careful selection of building materials and products. Today a number of low and no VOC building materials are available, including less polluting paints, adhesives, solvents, cleaning agents, caulking, wood products, carpets and sealants. Choose those products that have been tested and verified by reputable third-party certification programs like Green Seal and Green Guard. This will ensure that products have been tested and found to conform to the most stringent standards for low impact on the indoor environment.

The EPA recommends using IAQ-safe products that are non-toxic and not prone to moisture damage or mould growth. Proper housekeeping practices should be followed to keep dust levels low and uses third party-certified environmentally-friendly cleaning products, which will not negatively affect indoor air as much as chemically-laden counterparts.

1.6. IAQ management via modelling

The goal of air quality management is to maintain a quality of air that protects human health and welfare. This goal recognises that air quality must be maintained at levels that protect human health, but must also provide protection of animals, plants (crops, forests and natural vegetation), ecosystems, materials and aesthetics, such as natural levels of visibility (Murray 1997).

Ventilation based systems are usually most efficient, and coupled with the recent progress in control systems it may currently be some of the most effective solutions. Current algorithms of heating, ventilation, and air conditioning rely on control algorithms based on real-time sensing of control parameters. Various modelling approaches have been utilised to predict and manage such systems, including white box, black box, and grey box (ASHRAE 2009 and Afram and Sharifi, 2014a). White box models are based purely on the deterministic equations representing physical processes. If these are not entirely known (which is usually the case with complex systems such as indoor air), the relationships between input and output signals is approximated using experimental data (measured input and output signal values). In grey box models, analytical solutions to equations of the processes are known while experimental data is used only for determination of some parameters for analytical solutions. The above modelling approaches have been transferred to applications such as sub-zonal and multi-zone models, Computational

Fluid Dynamics, and a Windows-based IAQ simulation (Guo, 2000; Kassomenos et al., 2008 and Ren and Stewart, 2005).

The modelling of HVAC systems usually prioritise energy saving since HVAC systems use in average 40% of household energy (Afram and Sharifi, 2015). Currently, HVAC systems are mostly managed using three parameters: temperature, relative humidity, and carbon dioxide (CO_2) concentrations. The first two assure thermal comfort of residents while the CO_2 concentration represents IAQ as polluted by humans (Nielsen and Drivsholm, 2010 and Emmerich and Persily, 1997). CO_2-based measurement systems are mostly employed in public premises having large amounts of humans in closed environments.

Some studies investigated whether controlling ventilation by measuring occupancy (CO_2-based HVAC) could keep pollutants (e.g., formaldehyde, TVOC, radon) below their reference or regulatory limits (Chao and Hu 2004 and Herberger and Ulmer 2012). Some studies found that HVAC was not sufficient to control the measured pollutants below their established limits; additional studies would have found that HVAC did not control these pollutants if they considered the current reference exposure or standard limits for these pollutants. In residential environments, the range of indoor pollutants is much broader and does not depend only on the presence of the human body. HVAC-based CO_2 does not control outdoor generated pollutants, nor does it account for pollutants generated indoors but independently of human activities. Thus, the ability of the ventilation rate or HVAC to maintain an acceptable IAQ in buildings strongly depends on the source strengths, pollutant sources and infiltration rates, which are specific to building type and location.

Carcinogenic radon gas, which originates from rock and soil has been utilised as one of parameters for controlling HVAC (Chao and Hu, 2004). While it is widely acknowledged that VOCs and PM are important indoor pollutants, the reports on the IAQ management modelling/algorithms based on real-time sensing of these pollutants are as yet been scares (Kim et al., 2014 and Kolarik, 2012). Many studies have researched the effects of various ventilation patterns to the PM concentrations indoors (Ai et al., 2015; Fisher et al., 2015 and Heudorf et al., 2009) but not vice versa.

1.7. The findings of the literature review

1. IAQ is one of the most important factors influencing the indoor microclimate. Air pollutants in buildings are linked to indoor building factors (structure, fabrics, coating, furnishing, ventilation system, etc.), the specific activities of the occupants, or to other sources found outdoors.
2. The biggest negative impact on IAQ is caused by active pollution sources, which in a relatively short time high PM and VOCs seriously degrades the IAQ.
3. PM and VOCs are specific and most important pollutants affecting human health indoors. Particle size is directly related to its potential for causing health problems. Small particles of less than 2.5 µm in diameter can be inhaled deeper into the lungs and have been associated with various fatal and non-fatal

cardiovascular diseases. As with other chemicals, the effects of VOC exposure depend on several factors including the type of VOC, the amount of VOC and the length of time a person is exposed. Each chemical has its own toxicity and potential for causing different health effects. VOCs in the indoor environment can contribute to SBS. It is very important to identify and control the levels of PM and VOCs in the indoor environment with the aim of ensuring a healthy living.

4. Low energy buildings typically use high levels of insulation, energy efficient windows, low levels of air infiltration and heat recovery ventilation to lower heating and cooling energy. The removal of pollutants via ventilation is being challenged due to the increasing occurrence of energy efficient buildings due to increasing demand in energy saving. Tightness of buildings and presumably decreased ventilation may have a negative impact on IAQ. The summary of the literature review confirmed that IAQ in low-energy buildings is inadequate.

5. Good IAQ may be achieved by multiple measures, among which is ventilation, air cleaning and prevention (using low emitting building materials, avoiding combustion sources indoors, etc.). The summary of the literature review confirmed that in order to maintain good IAQ and energy efficiency air cleaning technology should be used widely. For good IAQ PAC and in-duct air cleaners, which effectively removes contaminants from the indoor air are used most often.

6. Currently, IAQ management is conducted using CO_2 concentrations, which represents the human metabolic products, but does not reflect the activities carried out in the room.

2. METHODS AND MEASUREMENTS

The methods of the thesis were divided into four stages: IAQ in low energy buildings, characterisation of active indoor pollution sources, IAQ management by combined ventilation and PAC and a grey model to IAQ management in rooms based on real-time sensing of PM and VOCs. A detailed illustration of the stages of the doctoral thesis is presented in **Figure 2.1**.
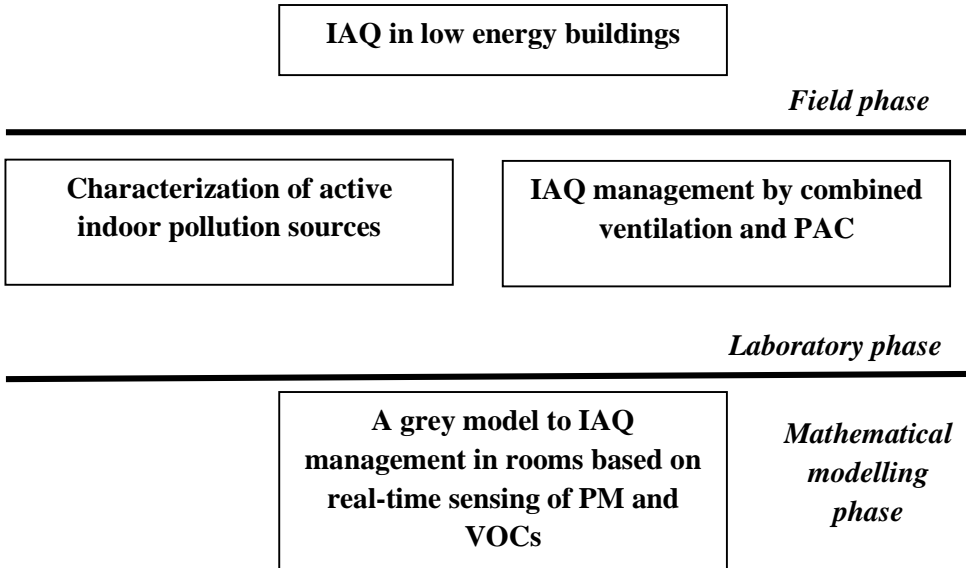
Figure 2.1. Stages of the doctoral thesis.

2.1. IAQ in low energy house

Measurement sites

The IAQ measurements were carried out in 11 single-family detached low energy buildings located in Kaunas (8 buildings) and Vilnius (3 buildings). The first round of measurements was performed during the period from April to August 2014. The building age of most of the buildings ranged from 2 months to one year, two unfinished buildings (B5 and B10) and one ten-year-old building (B8) were taken for comparison. Information about the sampling sites is summarised in **Table 2.1** it includes year built, energy efficiency class, useful area, type of ventilation and heating, furniture materials, occupancy and number of occupants. All buildings had a mechanical ventilation system installed and operating except for the two unoccupied buildings. It must be noted that although the mechanical ventilation systems were present, very few residents were aware of their running schedule or balancing.

Questionnaires and diaries were provided for the occupants. Questionnaires were constructed to collect information related to building construction characteristics and occupant activities during the measurement period. The main

data from the questionnaires used in these measurements, including background building information, are presented in **Table 2.1**.

To determine of the decrease of VOC concentration over time after construction the second round of the investigation was performed. The second round of the investigation was carried out in September 2014 in B5. The VOC concentration sampling was started the day after the complete installation of all interior surfaces and conducted over five consecutive weeks. In the measurement period the mechanical ventilation system was activated and ACR was kept constant at 0.5 h^{-1}. In this experiment was the building unoccupied.

Table 2.1. Description of the buildings investigated.

Parameter	Building number										
	1	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**	**10**	**11**
Year built	2013 05	2013 08	2013 08	2014 02	Not finished	2013 02	2013 05	2004	2011 01	Not finished	2014 02
Energy efficiency class	A+	A	A+	B	B	B	A	A	A	A+	A+
Useful area, m²	169	77	133	209	182	103	153	149	181	196	199
Type of ventilation	recuperative	recuperative	recuperative	recuperative	recuperative	recuperative	recuperative	recuperative	recuperative	recuperative	recuperative
Total primary energy use, kW/m²*a	40	129	34	109	61	107	63	26	98	49	49
Energy use for heating, kW/m²*a	17	65	15	83	33	77	38	17	55	23	24
Furniture	chipboard panels, solid wood, textile, linen	wood (polished), tapestry, leather	painted panels, cotton, tapestry, leather	wood, chipboard panels, tapestry	not installed	wood (polished), tapestry, faux, leather	chipboard panels, solid wood, textile, linen	wood (polished), tapestry, natural leather	wood, chipboard panels, tapestry	not installed	painted panels, cotton, natural stone
Occupancy	yes	yes	yes	yes	no	yes	yes	yes	yes	no	yes
Number of occupants	4	2	4	3	0	2	4	5	2	0	3
Area/resident ratio, m²	42	39	33	70	-	52	37	30	91	-	66

Measurements, sampling and chemical analysis

IAQ measurements in each building lasted one week (7 daily measurements). Indoor air samples were collected in the living room, or a hallway of the building. The measured gaseous pollutants indoors in the first round included formaldehyde, very volatile organic compound (VVOC), selected VOCs, semi-volatile organic compounds (16 US EPA defined PAHs, alkylated PAHs, PCBs, HCB), CO_2 and NO_2. Samplers of formaldehyde, VOCs and NO_2 were exposed for seven consecutive days. In the second round of sampling only concentrations of selected VOCs were measured.

A real time portable IAQ monitor (HD21AB IAQ Monitor, Delta OHM S.r.L., Italy), was used to monitor indoor CO and CO_2 concentration variances, as well temperature and relative humidity values. The sampling height was chosen according to the human breathing zone as seated, i.e. 1.2-1.5 m above ground. One-minute resolution was used. ACR was determined according to ASTM standard E741 – 11 using the decay of CO_2 concentration (ASTM E741 – 11). This procedure has been applied to the periods when residents noted full absence from the building.

Formaldehyde was collected with a Radiello passive air sampling system (Radiello, Fondazione Salvatore Maugeri, Italy). The Radiello passive air sampling system consists of the diffusive body, supporting plate, adapter and collection cartridge in the sealed glass tube. The sampling systems were positioned at a distance exceeding one meter from windows, door or other surfaces at a height of about 1.5–2.0 m above the floor. Formaldehyde sorbent cartridges matrix contains a 2.4–dinitrophenyl hydrazine coated Florisil adsorbent. After exposure, compounds were analysed with ultra-fast liquid chromatography coupled with UV/VIS and a diode matrix detector system (Prominence UFLC, Shimadzu, Japan).

The positioning and sampling procedure of VOC samplers was similar as for formaldehyde. VOCs sorbent cartridges contain a stainless steel net cylinder, with 100 mesh grid opening and 5.8 mm diameter, packed with 530±30 mg of activated charcoal with particle size 35-50 mesh (Radiello). VOCs are trapped by adsorption and recovered by carbon disulphide displacement. The samples were further analysed as follows: each sample sorbent was placed in a 4 ml vial; after placing the vial into the water bath, 2 ml of carbon disulphide was added (GC, Sigma Aldrich, USA) to each sample; the vial was tightly closed and put into the tube shaker. The duration of the extraction was 30 min. The extracted solvent samples were injected into the chromatographic vial by a 100 ml micro-syringe. The samples were analysed using a GC/MS system (GCMS-QP2010 Ultra, Shimadzu Corp., Japan) with a capillary column (Rxi-5ms, Restek Inc., USA). The temperature program was set to 30°C for 1 min, then raised at 40°C/min to 100°C (1 min), and finally at 1°C/min to 104°C, at which temperature it was held for 4 min. The MS scanning was performed from 30 to 450 m/s. The following target compounds were analysed: benzene, toluene, ethylbenzene and total xylenes.

NO_2 was collected using passive samplers (DIFRAM-100 – Rapid Air Monitor, Gradko International Ltd, UK) containing triethanolamine (TEA) as

the absorbent for seven consecutive days. Samples were analysed by Gradko, the laboratory was accredited by the United Kingdom Accreditation Service.

Gaseous SVOCs were sampled using semi-permeable membrane devices (SPMDs). SPMDs were deployed for one month in each location, thus representing a long term average pollutant level. SPMDs were attached to stainless steel holders. The analyses of the absorbed compounds were carried out by GC/MS system at the commercial laboratory ExposMeter AB, Sweden. The exposed membranes were dialyzed with two 250 ml portions of hexane (a more detailed procedure is presented in Alvarez et al., 2008). After the evaporation to 2-3 ml, the final volume of each dialysate was adjusted with n-heptane to 5 ml. 100 μl aliquots were mixed with 13C internal standards (2.5 ng per sample) in micro inserts of chromatographic vials, briefly sonicated, and forwarded for the analysis.16 US EPA PAHs and alkylated PAHs, PCBs and HCB were quantified. The levels of semi-volatile compounds determined by the passive method have been calculated in the accumulated mass per day of sampling (ng/day).

Quality control

The sampling, sample preparation, and analysis were performed according to the good laboratory management practice. Duplicate samples were taken in three sampling sites (27% of the total number). One transport blank was deployed in each sampling campaign. The analysis of the blank samples has not shown any contamination. Variations of concentration in duplicate samples did not exceed 15%. The estimated total cumulative uncertainty of the sampling and analysis methods was within 20%.

2.2. Characterisation of active indoor pollution sources

Tested indoor sources and the chamber

The choice of the pollution sources was based on the wide literature analysis of similar studies and classified into four main categories: cooking related sources (Hussein et al., 2006; Brauer et al., 2000 and Afshari et al., 2005), other thermal sources (Wu et al., 2012; Isaxon et al., 2013; Fine et al., 1999; Pagels et al., 2009 and Morawska and Zhang, 2002), personal care products (Nazaroff and Weschler, 2004 and Gehin et al., 2008), and household sources (Glytsos et al., 2010;Weschler and Nazaroff, 2008; Lioy et al., 1999 and Jones, 1999). The studied pollution sources and testing conditions are explicitly listed in **Table 2.2.**

Table 2.2. Detailed description of test conditions of indoor air pollution sources.

Indoor air pollution sources	Test conditions	Exposure time
Cooking related		
Cooking oil heating on electric stove	The pot (half a litre of oil) was heated on a heating plate (operating on maximum power).	15 min
Electric stove operating alone	Electric stove (electric power 1,500 W) operating for 20 min on maximum power.	20 min
Onion frying in oil in a Teflon pan on electric stove	Onion frying was carried using a Teflon pan with 20 ml of sunflower oil. The oil was heated for 5 min on the maximum heat. Then onion was sliced into small cubes (approx. 0.5 cm) and added to the pan with hot oil. Onion was fried and mixed continuously for another 5 min.	10 min
Teflon pan heating on electric stove	Teflon pan was placed on the medium size heating plate for 10 min (operating on maximum power).	10 min
Water boiling on electric stove	The pot (one litre of water) was heated on a heating plate (operating on maximum power) for 20 min.	20 min
Other thermal		
Candle (paraffin and perfumed) burning	Paraffin and perfumed candles burned for about 10 min. The candles were lit with a gas lighter.	10 min
Cigarette burning	One cigarette (Nicotine: 0.5 mg, Tar: 6 mg, CO: 7 mg) burned for about 1 min.	1 min
Incense stick burning	Incense stick burned for about 1 min. The incense was lit with a gas lighter.	1 min
Use of personal care products		
Hair dryer operating	Hair dryer (Philips, Salon Essential, 1,200 W) operated for 10 min. Two modes: medium hot air and hottest air.	10 min
Hair spray spraying	Hair spray spraying in the middle of the room for 5 seconds.	5 s
Household		
Air-freshener (aerosol) spraying	Spraying air-freshener in the middle of the room for 5 seconds.	5 s
Electric air heater operating	Electric air heater (Duracraft, DF 130E2, 2,000 W) operated for 10 min.	10 min
Electric radiator operating	Electric radiator (Heller MAB 2009, 2,000W) operated on the maximum power for 10 min.	10 min
Floor vacuuming	Vacuuming of the laboratory floor (covered with PVC layer) by a vacuum cleaner on the maximum power (Electrolux, Ultra silencer, Max. 1,250 W) lasted about 15 min.	15 min
Floor wet mopping with detergent	Wet mopping of the laboratory floor with detergent (floor cleaner "Ūla" for laminate flooring and linoleum) (covered with PVC layer) lasted about 5 min.	5 min
Furniture polisher spraying	Spraying furniture polisher in the middle of the room for 5 seconds.	5 s
Ironing of clothing	A flat iron with and without steam was used, respectively, on a cotton sheet, which was washed in warm water.	10 min

Simulations were carried out in a test chamber with the floor area of 13 m^2 and volume of a 35.8 m^3, representing a typical room, which was installed inside a 150 m^3 laboratory premises. The distance from the chamber floor to the air supply diffuser was 2.3 m and 2.8 m to air exhaust diffuser (equal to chamber height). The distance between the exhaust and supply diffusers was 2.6 m. **Figure 2.2** shows the schematic diagram of the experimental setup, where pollution monitors were position outside of the chamber. The walls, floor and ceiling of the chamber were fabricated using conventional construction materials, such as painted dry-wall, PVC floor lining, and a panel ceiling. The ventilation air was supplied via the air handling unit (Gold04, Swegon, Sweden), assuring the ACR of 0.5 ± 0.1 h^{-1} in the chamber in order to simulate typical ventilation conditions, either in forced or natural mode. The ACR of the test chamber volume was checked before and after each experiment via the air velocity measurements. Additionally, the ACR was verified using the CO_2 decay method (ASTM E741 – 11). The nozzle diffuser was built from 25 separate nozzles supplying the air downwards at 45° angle to the wall.
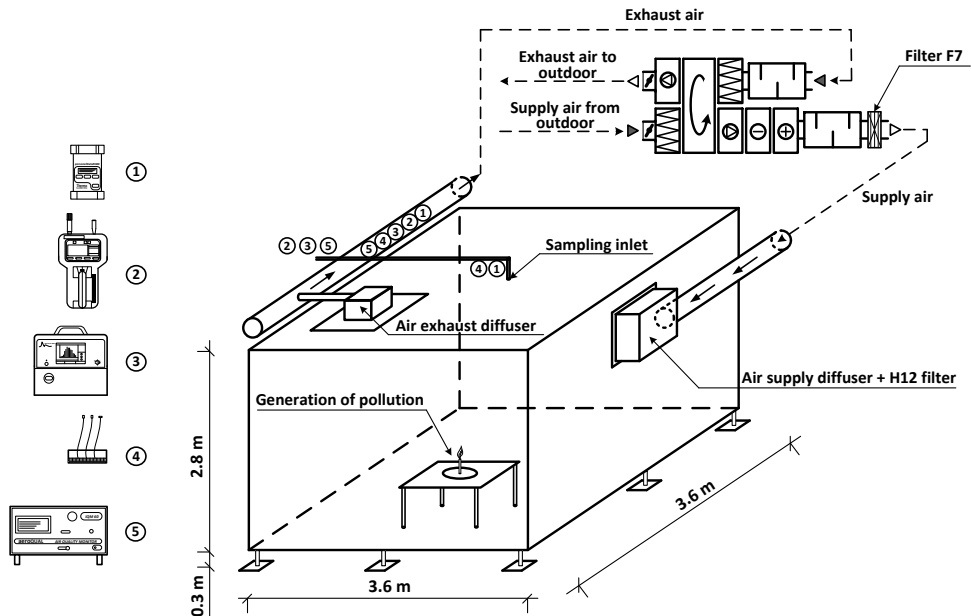
Figure 2.2. Test chamber with air supply and exhaust system via air handling unit with heat exchanger. 1 – Thermo Scientific pDR-1000AN monitor (PM10.0); 2 – Optical particle counter 3016 IAQ ($PM_{0.3-10.0}$); 3 – Scanning mobility particle sizer SMPS-3910 ($PM_{0.01-0.3}$); 4 – VOC/T/RH logger (VOC sensor – iAQ–2000); 5 – Indoor air quality monitor IQM–60 (CO, CO_2, O_3, NO_2, VOC, NMHC).

Low concentration of coarse PM (<300 p/cm^3) in the supply air was ensured using an F7 class filter (constructed inside the handling unit) and high efficiency particulate air filter (HEPA 12, General Filter, Italy) installed at the supply diffuser. The temperature (T) and relative humidity (RH) were recorded during the

experiments, and ranged from 18 to 22°C and 30-70% (water boiling experiments), accordingly.

The description of the modelling of each source with the tested procedure is presented in **Table 2.2**. After the end of each experiment, the room was purged with air until the concentrations of particles reached the background values (observed values before the initiation of the experiment). Before the start of each run a background concentration of PM was recorded (3 min before the start). Experiments for each source were repeated three times. A pollution episode was generated and the dynamics of particle concentrations were monitored for 30 min. This duration was selected due to the limitation of overall experiment time. Each run required approximately 90 min of purge of the chamber before the start of the experimental run, which lasted for 30 min. The analysis of the results has later indicated that in some cases this period has been insufficient to reduce the concentration of PM back to the background level.

Commercial computational fluid dynamics (CFD) code was used to predict contaminant dispersion and air flow patterns in the test chamber. The K-epsilon turbulence model, Cartesian grid of 400 thousand cells, and line-by-line tri-diagonal matrix (TDMA) algorithm with Double Precision Solver (DPS) was chosen for steady state numerical simulations. Two cases were analysed by means of CFD: warm and isothermal pollutant injection with the air temperature equal to +30°C and +20°C respectively. PM0.5 was used as the contaminant for both cases. The model of the nozzle diffuser was sketched keeping the same geometric shapes to the diffuser installed in the chamber.

In case of exhaust ventilation, measurements were carried out in the air duct, 1.5 m from the air diffuser to ensure minimised turbulence of the air flow due to the bends in the duct. Two isokinetic sampling probes were used to sample particles, as specified in the subsequent chapter. In the centre of the ceiling, sampling was conducted via Tygon tubes (inner diameter 10 mm, length 2 m) introduced in the centre of the ceiling. Obtained results were adjusted for diffusion losses, which ranged from 11% (particle size 0.01 μm) to 1% (0.1-0.3 μm) (Baron and Willeke, 2001).

Measurements

The specific experiment has been performed in order to test the effect of measurement locations to the variation of PM concentrations. Separate measurements for each source were performed in two places: in the exhaust ventilation and in the centre of the ceiling. It was hypothesised that exhaust air should provide an integrated parcel of polluted air in a well-defined volume and flow, while the centre of the ceiling should represent a faster response, but slower change in the concentration.

Instruments

A Scanning Mobility Particle Sizer (SMPS, model SMPS-3910, TSI Inc., USA) was used for measurements of the FP number size distribution within the 10

to 420 nm size range. The SMPS scanned across 13 size bins from 10 to 420 nm ($PM_{0.01-0.3}$) of electrical mobility diameter during 60 s, or was used as a counter for single size mode at 1 s resolution, with the flow rate of 1 l/min. The global normalised error was found at 0.55-0.97 comparing Nanoscan SMPS-3910 versus laboratory calibrated SMPS for indoor aerosols under laboratory conditions (Stabile et al., 2014).

An Optical Particle Counter (OPC, model 3016IAQ, Lighthouse worldwide solutions, Fremont, CA, USA) was used for the determination of PN size distribution within the 0.3 and >10 μm size range. The OPC measured particles in a range of 6 channels by optical diameter: >0.3, >0.5, >1.0, >2.5, >5.0 and >10.0 μm ($PM_{0.3-10}$) at the flow rate of 2.83 l/min.

The mass concentration of PM aerosol was measured by light-scattering photometer – nephelometer (pDR-1000AN, Thermo Fisher Scientific Inc., USA). A nephelometer operates passively, that is, it relies on diffusion of air into the particle sensing region such that particle concentrations can be quantified. Because it has no internal pump system to draw air through the sensing unit, it cannot segregate particles into size fractions before sensing. The nephelometer measures airborne particles with a diameter of <10 μm (PM10), with a concentration range of 0.001-400 mg and ±5% precision.

The IAQ Monitor (IQM-60, Aeroqual Limited, New Zealand) detected VOCs via a PID and MOS sensor (the latter referred to as a gas sensitive semiconductor (GSS) sensor by the manufacturer). IQM-60 provides concentration readings in ppm of VOC concentration based on the calibration isobutylene with every 120 seconds. The measurement range was 0-20 ppm for PID and 0-25 ppm from MOS, and detection limit 0.01 ppm and 0.1 ppm, respectively. IQM-60 also measured ozone at a measurement range of 0-0.5 ppm for O_3 and a detection limit of 0.001 ppm.

Concentrations of VOCs were measured using an air quality sensor based on micro-machined metal oxide semiconductor (MOS) technology (iAQ2000, AMS Sensor Solutions Germany GmbH). The metres are sensitive to various non-methane hydrocarbons, including aliphatic hydrocarbons and aromatic hydrocarbons, alcohols, ketones, organic acids and amines, as well as methane. The quantitative response of these sensors was obtained by using manufacturer provided calibration based on the Reversed Metabolic Rule technology, which calibrates measured VOC concentrations to CO_2-equivalent ppm values, thereby achieving compatibility to CO_2 standards (Herberger et al., 2010).

Data processing and analysis

Data processing and management was carried out using Excel 2010 (Microsoft Inc., USA) and OriginPro 9.0 (Version 9.0, OriginLab Corporation, USA) software. Values are expressed as mean ± standard deviation (SD). The Mann-Whitney rank sum test was used to compare measurements in different locations of the room. Differences resulting in *p*-values <0.05 were considered as statistically significant.

Mathematical representation of the dynamics of pollutant concentration

The dynamic pattern of the pollution sources was split into two phases: the growth of concentration and the decay of concentration. The growth phase statistically fit with a logistic three-parametric sigmoidal (S-shaped) function (**Figure 2.3**):

$$C_t = (C_{max} - C_{min})/(1 + e^{-\left(\frac{t-t_0}{b}\right)}) \qquad (1)$$

In which t denotes time (s), y is the PNC in the time t (p/cm^3); C_{max} - C_{min} is the interval of the upper asymptote minus lower asymptote; t_0 is the inflection point, i.e., time when maximum PNC is equal to the half of the interval (C_{max} - C_{min}) (s); b is the slope coefficient, representing the rate of change or slope at the inflection point x_0. Smaller b values represent quickly responding processes (e.g., fast increase in concentration of pollutants), while larger b values denote slower responses.

PNC decay rates generally represent the interaction between ventilation, gravitational settling, diffusion, electrostatic effects, and thermophoresis. Among these, the removal of PM due to ventilation and deposition is the most important and can be described by the solution of the first order differential mass balance equation (Abt et al., 2000 and He et al., 2005):

$$C_{(t)} = C_{(0)}e^{-(a+k)t} \qquad (2)$$

Where k is the particle deposition rate, a is the ACR, t is the interval time and C_t and C_0 are particle concentrations (p/cm^3) measured at times t and t_0, respectively.

In the case of our experiments, in order to more accurately represent the dynamic patterns of the decay, the process has been split into two phases (fast and slow). The logistic double four-parameter exponential decay function was used to characterise the decrease in pollutant concentration:

$$C_{(t)} = C_{(1)}e^{-d_1 t} + C_{(2)}e^{-d_2 t} \qquad (3)$$

In which t is time (s), y is the PNC in the time t (p/cm^3), C_1 and C_2 are the "amplitudes" of the individual exponential components, and d_1 and d_2 are the corresponding exponential constants (the decay rate of PNC) (**Figure 2.3**). The goodness-of-fit of the exponential regression curve was at least 90% ($R^2 > 0.9$).
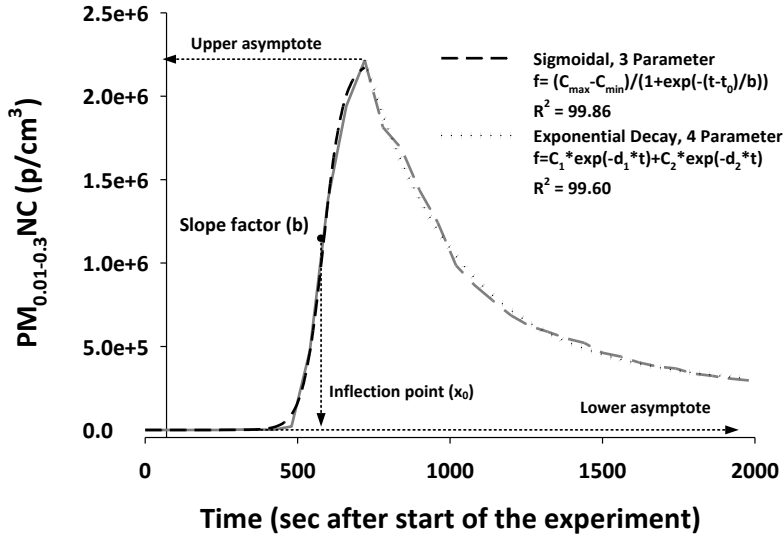
Figure 2.3. Characterisation of PNC curve by three–parameter sigmoidal (increase in concentration) and double four-parameter exponential decay equations (concentration decay) under the operation of cooking oil heating on electric stove.

Comparison of the effects of measurement location on the dynamics of pollution sources

The following parameters were used in order to assess the effect of the measurement location on the representation of the dynamics of the pollution sources: ΔC – number concentration difference between maximum observed PNC; Δt – difference between response time (i.e., time of travel of air parcel containing pollutants to the measurement location); the growth of concentration (based on a sigmoidal function, as described in the previous section); concentration decay rates (based on an exponential decay function). The concentration difference between maximum PNC levels in the two measurement locations was calculated according to the following equation:

$$\Delta C = |C_E - C_C| \tag{4}$$

In which C_E is the maximum PNC measured in the exhaust channel and C_C is the maximum PNC measured in the centre of the ceiling.

The response time difference between $PM_{0.01-0.3}$ response times in two measurement locations was calculated according following equation:

$$\Delta t = |t_E - t_C| \tag{5}$$

In which t_E is the PNC detection time in the exhaust channel and t_C is the FPs detection time in the centre of the ceiling.

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2.3. IAQ management by combined ventilation and air cleaning

Test chamber and a source of pollutants

The experiment was conducted in a test chamber representing a typical room, all detailed information is presented in the previous section. **Figure 2.4** shows the schematic diagram of the experimental setup. Sampling probes were positioned in the breathing zone of a seated person, 1.25 m above the floor adjacent to the opposite wall of a PAC, where pollution monitors were position outside of the chamber.
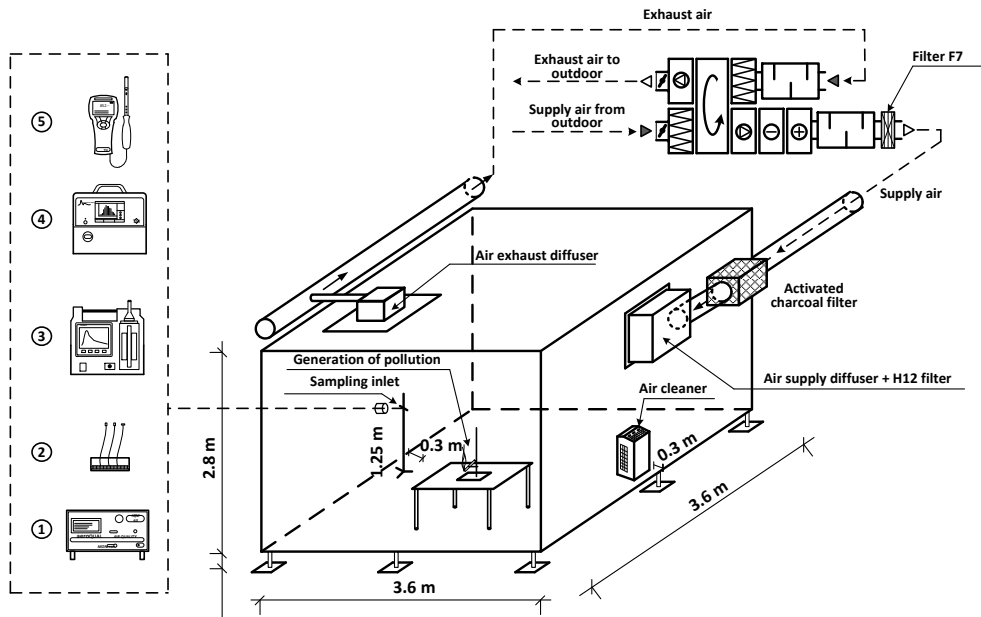
Figure 2.4. Test chamber with air supply and exhaust system via air handling unit with heat exchanger. 1 – Indoor air quality monitor IQM-60 (CO, CO_2, O_3, NO_2, VOC, NMHC); 2 – VOC/T/RH logger (VOC sensor – iAQ–2000); 3 – Electric low pressure impactor ELPI+ ($PM_{0.006-10.0}$); 4 – Scanning mobility particle sizer SMPS-3910 ($PM_{0.01-0.3}$); 5 – CO_2 monitor 7545 IAQ–CALC.

Cigarette smoke was utilised as a surrogate source of air pollutants. It is a well-known indoor air pollutant with adverse health effects, therefore used in PAC testing standards (AHAM, 2006 and JEMA, 1995). Cigarette smoke contains aerosol particles and VOCs (including acetone, 2,3-butanedione, 2-butanone, benzene, ethylbenzene, styrene, toluene, etc. (Polzin et al., 2007)), making it suitable for co-testing PAC performance both on solid and gaseous phase pollutants. A cigarette having a declared yield of nicotine (0.5 mg), tar (6 mg), and CO (7 mg) was placed in a holder on a table inside the chamber and burned for about 10 min. Background IAQ was monitored for three minutes before the start of each experiment. The variation of pollutant concentrations due to the ventilation and air cleaning was monitored for 90 minutes. The chamber was moped with cleanser-free tap water and allowed to dry overnight after each day of experiments.

Ventilation

The ventilation process represented a single-pass filtered outdoor air, supplied and exhausted via the air handling unit (GOLD 04, Swegon AB, Sweden). During the experiments the supply air temperature was set to +20ºC. The temperature and relative humidity of the air ranged between 18-19ºC and 38-49% in the chamber during the experiments, respectively. Low concentrations of particles in the supply air was ensured using an F7 class (adequate to MERV13) filter (constructed inside the handling unit) and subsequently a high efficiency particulate air filter (HEPA 12, General Filter, Italy) installed at the supply diffuser mounted on the wall close to the ceiling (**Figure 2.4**). The gaseous organic pollutants were removed by a fixed active carbon bed. Air was exhausted through a single port located in the ceiling in the opposite side of the chamber.

ACR inside of the chamber was checked before and after of each experiment via the duct air velocity measurements. Additionally, the ACR was verified using the CO_2 tracer gas decay method (ASTM E741 – 11) by CO_2 meter (7545 IAQ–CALC, TSI Inc., USA).

Air Cleaning

The air cleaning process was represented by multi-pass air treatment by PAC. Three commercially available PAC (C1, C2, and C3, **Table 2.3.**) have been tested. All units were designed to serve for similar areas (46-48 m^2) and were rather equally priced. All units operated a fan to draw the air through a combination of particle phase and gaseous phase pollutant removing stages. The first cleaner (C1) passed the air through three filters (a pre filter, a washable activated carbon filter, and an HEPA 13 equivalent filter), followed by "Plasmacluster ion technology", which is declared to deactivate the suspended airborne bioaerosol and decompose odour molecules. The C2 had a complex set of six-stage pollutant removal sections, including a streamer discharge unit, pre filter, plasma ioniser, electrostatic filter, titanium apatite (photo catalyst), and deodorising catalyst filters. The C3 passed the air through a "micro filter" (removing particles >0.4 µm), oxygen-activating module (ozone based), and a catalyst.

The PACs were placed inside the test chamber in accordance with the air cleaner installation guidelines, i.e., on a floor below the air supply diffusers. The PAC was activated at the same time as the source of pollution.

Table 2.3. Specifications of the tested PAC as provided by the manufacturers.

Device	Manufacturer	Model	Dimension, mm	Approx. Price, €	Applicable area, m²	Technology
C1	Sharp Corporation, Japan	KC–A60	643 H × 416 W × 295 D	500	48	Pre filter, washable activated carbon filter, HEPA 13 filter, "Plasmacluster technology".
C2	Daikin, Japan	MC70L	576 H × 403 W × 241 D	575	46	Streamer discharge, pre filter, plasma ioniser, electrostatic dust collection filter, titanium apatite filter, deodorising catalyst filter.
C3	Zepter International, Germany	Therapy Air	400 H × 515 W × 140 D	575	46	Pre filter, "micro filter", oxygen activating module, catalyst.

IAQ indicators and monitors

The following physical and chemical parameters of indoor air were selected as proxy parameters to represent the efficiency of ventilation and PAC: PNC and concentration of VOCs.

The size-resolved decay of PNC was measured using a SMPS 3910 (all detailed information is presented in the previous section) and an Electric Low Pressure Impactor (ELPI+, Dekati Inc., Finland). The ELPI+ divided aerosol particles to 15 fractions (0.006–10.0 µm) sampling at a flow rate of 10 lpm. This instrument utilises the cascade impaction principle and also has a direct-reading capability. Aerosol samples were collected on 25 mm diameter aluminium foil substrates covered with Apiezon L grease. Real-time concentrations of aerosol samples were registered in one-second intervals. Aerodynamic diameter data obtained by ELPI+ was converted to mobility diameter using cigarette smoke density of 1,180±113 kg/m³ (Johnson et al., 2014 and Khlystov et al., 2004).

The concentration of total gaseous organic compounds was measured using several instruments operating photo ionisation detector (PID) and metal oxide semiconductor (MOS) sensors. These technologies are some of the widest spread currently for the measurement of real-time total organic compound concentrations, although having some differences in responses to various substances, selectivity, and accuracy. The IQM–60 monitor (Aeroqual Limited, New Zealand) detected VOCs via the PID and MOS sensor (the latter referred to as a gas sensitive semiconductor (GSS) sensor by the manufacturer) (all detailed information is presented in the previous section). With the aim of obtaining a higher temporal resolution, another

MOS sensor (iAQ2000, AMS AG, Germany) (all detailed information is presented in the previous section) was included.

The CO_2 monitor (7545 IAQ-CALC) and the VOC sensor (iAQ-2000) together with temperature and relative humidity sensors (PT907, Pace Scientific, USA) were installed inside the chamber, approx. 1.25 m above the floor. The IQM–60 and PM monitors were positioned outside the chamber and the air samples have been delivered via 0.5 m long Tygon tubes of 6 mm in diameter. The obtained results were adjusted for diffusion losses, which ranged from 3% (particle size 0.01 µm) to <1% (0.1–0.3 µm) (Baron and Willeke, 2001). The electricity consumption of the air cleaners was monitored using an energy meter (EL-EPM02HQ, Nedis B.V, Netherlands).

Main parameters

The concentration decay rate was calculated following a first-order decay model:

$$C_t = C_0 e^{-kt} \tag{6}$$

Where C_t is the concentration at time t (p/cm^3 or ppm), C_0 is the initial concentration at t = 0 (p/cm^3 or ppm), k is the concentration decay rate (h^{-1}), t is the time (h).

The pollutant removal efficiency was calculated as follows (JEMA, 1995 and KACA, 2006):

$$E_P = \left(1 - C_t / C_0\right) \times 100 \tag{7}$$

Where E_P is the pollutant removal efficiency (%, further represented as E_{PNC} and E_{VOC}), C_0 is the initial concentration at t = 0 (p/cm^3 or ppm), C_t is the concentration at time t = 0.5 h (p/cm^3 or ppm).

The CADR was calculated using the method by AHAM (2006), additionally subtracting concentration decay due to pollutant loss in the chamber:

$$CADR_P = V(k_e - k_n - k_c) \tag{8}$$

Where $CADR_P$ is the clean air delivery rate (m^{3}h^{-1}) for a certain pollutant (further represented as $CADR_{PNC}$ and $CADR_{VOC}$), V is the volume of the chamber (m^3), k_e is the total decay rate, including both cleaner and ventilation (h^{-1}), k_n is the natural decay rate, including only ventilation (h^{-1}), k_c is decay rate of the pollutant concentration, reflecting loss of pollutants due to deposition or adsorption (h^{-1}), which was indicated as an important parameter by (Mølgaard et al., 2014).

The CADR was based on the concentration decay slopes with R^2 value above 0.97 derived from 30 data points (0.5 h of decay). CADR was calculated for particles up to 1.2 µm, since cigarette smoke did not generate sufficient amounts (>100 p/cm^3) of larger particles.

It must be pointed out that the chamber in which the experiments were performed did not meet the standardised requirements for the chamber volume and

surfaces. PAC testing was performed using additional air mixing with a fan, while combined ventilation and air cleaning experiments were performed with both full mixing and no additional mixing. It was found that there was a good agreement between well-mixed and no mixing conditions, possibly due to the fact that PAC provide substantial air flow to facilitate mixing of pollutants in the room.

The EEI was calculated based on the electrical power drawn for both air handling unit and PAC. The power draw of PAC was directly measured, while that of the air handling unit was estimated based on the operating time, flow rate and a specific fan-power of 2 kWsm^{-3} for the ventilation with a heat recovery (Nilsson, 1995). The EEI was computed with the following equation:

$$EEI_P = CADR_P / OP \qquad (9)$$

Where EEI_P is the energy efficiency performance index for a certain pollutant (further represented as EEI_{PNC} and EEI_{VOC}), $CADR_P$ is the clean air delivery rate (m^{3}h^{-1}) and OP is the electrical operating power of the PAC (W).

Experimental design

The study was designed as a two-phase experiment. During the first phase three PAC were characterised in the test chamber under the minimum mechanical ventilation conditions according to their flow rate (min and max modes), particle and VOC removal efficiency (E$_{PNC}$ and E$_{VOC}$), CADR based on particle and VOC concentration decay (CADR$_{PNC}$ and CADR$_{VOC}$), power draw, and energy efficiency performance index (EEI$_{PNC}$ and EEI$_{VOC}$). All experiments were repeated three times to obtain an estimate of a random error.

During the second phase, the combined effects of ventilation and PAC to the IAQ were analysed in a frame of controlled experiment, designed and fitted to regression models with the aid of a software package (Modde 10, MKS Umetrics, Sweden). Experimental variables included mechanical ventilation intensity (0.2 h^{-1}; 0.6 h^{-1}; 1.0 h^{-1}) and PAC device regime (min and max modes) expressed as relative air changes per hour (h^{-1}), i.e. cleaner flowrate divided by the chamber volume. A full factorial experiment with three middle points was conducted. Twenty-two experiments were performed to obtain the response parameters, including the concentration decay rate (k), CADR$_{PNC}$, CADR$_{VOC}$, E$_{PNC}$, E$_{VOC}$, EEI$_{PNC}$, and EEI$_{VOC}$. The experimental data was then fitted to the partial least squares regression model in order to obtain polynomial function based response surface plots, relating experimental variables and responses.

Data processing and analysis

Data analysis was performed using SSPS 13 (IBM Corp., USA) and Origin 9 (OriginLab Corp., USA) software packages. Every experimental result was reported as the average with the corresponding standard deviation provided where appropriate.

2.4. A grey model to IAQ management in rooms based on real-time sensing of particles and VOC

Design of the study

The study has been fulfilled through several phases which are presented in forthcoming chapters: a) Experimental phase, where the temporal variation of pollutant concentration has been investigated during various pollution episodes in a controlled environment; b) mathematical modelling phase, where the data on pollutant variation has been applied to the indoor pollutant mass balance model, with the aim of investigating the effects of ventilation rate to real-time reduction of pollutant concentration in a room.

Experimental phase

The experimental portion and all detailed information is presented in the previous section 2.2. Characterisation of active indoor pollution sources.

IAQ model

Further analysis of data was based on the IAQ mass balance model (Hussein et al., 2008). Such models are widely applied to describing effects of pollutant generation within the room due to pollution sources or with ventilation air, as well as removal due to ventilation, deposition and other removal mechanisms.

The general equation for the model is presented as follows:

$$\frac{d}{dt}N_p = \lambda P_i N_O - (\lambda + \lambda_d)N_p + S \tag{10}$$

Here N_p is the pollutant concentration in a room (ppm for VOC, mg/m^3 for coarse PM, #/cm^3 for fine PM), t is time (s), N_O is pollutant concentration in the outdoor (ventilation supply) air (ppm for VOC, mg/m^3 for coarse PM, #/cm^3 for fine PM), P_i supply air flowrate (m^3), λ is air change rate (s^{-1}), λ_d is particle deposition rate (s^{-1}), S is pollution source intensity (ppm/s for VOC, mg/m^{3}s for coarse PM, #/cm^{3}s for fine PM).

In the case of this experiment, several assumptions were made. The supply ventilation air was assumed to be pollutant free (which was achieved by using HEPA filtration). In the case of ventilation ACR was equal to or higher than 0.5 h^{-1}, $\lambda_d \ll \lambda$. Thus Equation (10) may be reduced as follows:

$$\frac{dN_p}{dt} = -\lambda N_p + S \tag{11}$$

Usually it is assumed that the emission rate of pollutants from a pollution source is constant in time (Hussein et al., 2006). In such case the solution to the equation (concentration of pollutant in a room at time t) is as follows:

$$N_p = \frac{S}{\lambda}\left(1 - e^{-\lambda t}\right) \tag{12}$$

At the same time, such assumption of constant pollutant emission is not always valid in real life scenarios. The emission rate depends on the process during which pollutants are being generated. Moreover, in order to apply IAQ management in a room, the ventilation rate must be changed ($\lambda \neq$ Const.). In such scenarios, equation (11) is presented in the following format:

$$\frac{dN_p}{dt} + \lambda(t)N = S(t) \tag{13}$$

This equation has served as the basis equation for the modelling within this study. The Simulink algorithm is presented in annex material (**Annex 5A**). The solution of this equation requires the knowledge of the variation of the pollutants source strength in time. This data is not available, thus we have derived it from the measurements of the pollutant concentration variation in the chamber experiments at a constant air change rate λ, as described in Ciuzas et al. (2015). This variation of pollutant concentration is approximated by a function $f(t)$:

$$f(t) = \frac{0.5a_0}{1 + \exp\frac{t_1 - t}{b_0}} (\mathrm{sgn}(t_1 - t) + 1) +$$

$$\sum_{i=2} a_i \exp(-b_i(t - t_{i-1})) \frac{(\mathrm{sgn}(t - t_{i-1}) + 1)(\mathrm{sgn}(1 - (t_i - t)))}{4} \tag{14}$$

Here a_0 and b_0 are parameters of sigmoidal function, representing an increase in concentration, a_i and b_i are parameters of exponential function representing the decrease in concentration.

The Equation (14) describes all pollution sources that operate in increase/decrease regimes. For those sources which exhibit multiple stages of increasing emissions (i.e., candle burning with an increased emission after extinguishment of a flame), the Equation (14) may be amended by additional (up to five) members, each representing an additional stage of increase/decrease:

$$f(t) = \frac{0.5a_0}{1 + \exp\frac{t_1 - t}{b_0}} (\mathrm{sgn}(t_1 - t) + 1) +$$

$$\sum_{j=2} \sum_{i=2} a_{ij} \exp(-b_{ij}(t - t_j)) \frac{(\mathrm{sgn}(t - t_{j-1}) + 1)(\mathrm{sgn}(1 - (t - t_j)))}{4} \tag{15}$$

The source strength $S(t)$ can be calculated by solving the equation:

$$S(t) = \frac{dN_p}{dt} + \lambda(t)N_p = \frac{df(t)}{dt} + \lambda(t)f(t) \tag{16}$$

Such equation can be easily solved using the algorithm, presented in **Annex 5B**.

The function $f(t)$ represents the decrease of concentration and varies accordingly:

$$f(t) = a_1 e^{\alpha_1 t} + a_2 e^{\alpha_2 t} \tag{17}$$

Hence Equation (13) is solved as follows:

$$S(t) = (\lambda - \alpha_1) a_1 e^{-\alpha_1 t} + (\lambda - \alpha_2) a_2 e^{-\alpha_2 t} \tag{18}$$

In cases when $\alpha_1 > \lambda$ or $\alpha_2 > \lambda$, additional pollutant removal mechanisms (such as particle deposition on surfaces) are working together with the removal with ventilation. The Equation (18) is amended with additional factor:

$$\frac{d}{dt} N_p = -(\lambda + \lambda_d) N_p + S(t), \tag{19}$$

Practically a minimum value of λ_d is applied so that the source $S(t)$ would be non-negative. The calculation algorithm is amended accordingly, as displayed in **Annex 5C**.

3. RESULTS AND DISCUSSION

3.1. IAQ in low energy buildings in Lithuania

3.1.1. Indoor microclimate

The relative humidity (40-60%) in the buildings fell within the comfort range as defined by the Lithuanian guideline values (HN 42:2009) (**Table 3.1**). The relatively small variation in RH was favourable for VOC measurements, which have been shown to be affected by low RH values (2-3-fold lower concentrations of some VOCs at lower RH levels (RH 21-22% compared to at RH 58-75%) (Markowicz and Larsson, 2015). The thermal conditions, however, where not always comfortable. The absolute maximum registered temperature has not exceeded the comfort temperature of 25°C only in three tested energy-efficient buildings. Temperatures above 27°C were registered in B6, B10, and B11. In B6 and B10, 25 and 26 degree hours above 27°C were observed during the one-month measurement campaign, while in B11 the overheating burden was the highest, where 224 degree hours above 27°C were observed. This illustrates that envelopes of tested energy-efficient buildings cannot successfully prevent indoor air from possible overheating during warm seasons, once outdoor temperature exceeds 30°C (in some cases outdoor temperature was observed around 34–35°C). Although Lithuania is often considered as a part of Northern Europe, the changing climate raises challenges for energy-efficient buildings to perform not only in low temperature but also high temperature conditions. Shading of building windows/envelope from the direct sunlight should be considered as one of the measures trying to prevent indoor air from overheating during the warm season.

The estimated ACR in most of the buildings rate was low (median 0.2 h^{-1}), ranging from 0.08 to 0.69 h^{-1} (**Table 3.1**). Such results indicate potentially insufficient ventilation conditions in most of the researched buildings, possibly due to a lack of professional setup after installation, or inadequate knowledge in operation/management by residents. Earlier research has also indicated cases of low ventilation in low energy buildings, i.e. Derbez et al. (2014) reported that in such buildings adjustment of the ventilation system to low settings might lead to an ACR as low as 0.1-0.2 h^{-1} (Derbez et al., 2014). Low ACRs were also observed in zero-energy buildings in Massachusetts as concluded by the authors because of the lack of information about "proper" ventilation for occupants and for the builder (Engelmann et al., 2014). Although insufficient ventilation rates indicated a potential to accumulate CO_2 indoors, the CO_2 levels were mostly within recommended threshold values. The mean concentration of CO_2 among the buildings ranged from 439 ppm (B5) to 1,117 ppm (B2). In B2 the concentration of CO_2 exceeded 1,000 ppm 35% of the measured time, and twice exceeded 2,000 ppm. The increase CO_2 values occurred during the nigh-time, since residents were switching the ventilation off due to uncomfortable airflows in their sleeping area. The exceedance of 1,000 ppm was very episodic in other buildings (1-3% of the measurement time). Although the ACR was low, we attribute low CO_2 levels to a high area/resident ratio in most of the buildings (ranging from 30 to 91 m^2/person) (**Table 3.1**), which allowed sufficient amounts of air to dilute exhaled CO_2. The tested buildings were relatively large as indicated by the area (**Table 3.1**); it appears that in Lithuania low energy is not associated with the reduced size of single family homes.

Table 3.2. The summary of measured microclimate and IAQ parameters in 11 low energy buildings.

Parameter	Limit value*	1	2	3	4	5	6	7	8	9	10	11	Range	Mean
T, °C		21	21.6	22.1	20.6	16.2	23.4	23.4	23.9	23.8	24	23.8	**16.2-24.0**	**22.2**
RH, %		51	44	50	51	50	48	52	52	51	58	55	**44-58**	**51**
CO_2, ppm		655	1117	562	765	439	636	845	691	572	451	669	**439-1117**	**672.9**
ACH, h^{-1}		0.11	0.21	0.11	0.69	0.09	0.39	0.26	0.37	0.12	0.08	0.15	**0.08-0.69**	**0.2**
NO_2, $\mu g/m^3$	**40**	4.6	2.8	4	5.1	1.2	5.3	5.3	1.9	3.1	1	5.4	**1.0-5.4**	**3.6**
Benzene, $\mu g/m^3$	**100**	0.8	0.8	0.6	8.8	6.4	0.6	2.3	1.3	1.2	0.2	0.3	**0.2-8.8**	**2.1**
Ethylbenzene, $\mu g/m^3$	**20**	2.1	1.4	1.1	521.7	8.4	0.1	1.1	1.1	0.5	0.2	0.8	**0.1-521.7**	**49**
Toluene, $\mu g/m^3$	**600**	5.2	2.8	4.1	18.5	33.6	3.8	8	1.4	4.2	0.1	1.9	**0.1-33.6**	**7.6**
o-Xylene, $\mu g/m^3$	**200****	0.9	0.4	0.2	328.9	6.5	0.2	0.9	0.4	0.5	0.1	1.1	**0.1-328.9**	**30.9**
p-Xylene, $\mu g/m^3$		0.9	0.3	0.5	214.5	11.6	0.3	1.5	1.3	0.9	1.1	1.4	**0.3-214.5**	**21.3**
BTEX, $\mu g/m^3$		**10**	**5.8**	**6.5**	**1092.4**	**66.4**	**5**	**13.8**	**5.5**	**7.3**	**1.8**	**5.5**	**1.8-1092.4**	**110.9**
Styrene, $\mu g/m^3$	**2**	0.3	0.4	0.3	419.8	0.7	0.1	0.5	0.1	0.1	0.9	1	**0.1-419.8**	**38.6**
Trichloroethylene, $\mu g/m^3$	**1000**	5.5	8.2	7.6	29.7	21.9	7.4	7	5.6	6.2	0.1	0.7	**0.1-29.7**	**9.1**
Tetrachloroethylene, $\mu g/m^3$	**60**	1.1	1.7	1.1	4.5	2.8	0.9	1.1	1.2	1	0.5	2.8	**0.5-4.5**	**1.7**
Formaldehyde, $\mu g/m^3$	**10**	17.5	30.8	20.8	37.1	28.8	40.5	52.3	37.5	26.7	3.3	42.2	**3.3-52.3**	**30.7**
Sum PAHs, ng/SPMDxday^{-1}		43	57	79	86	94	56	31	44	34	30	35	**29.7-94.3**	**53.5**
Sum alkyl-PAHs, ng/SPMDxday^{-1}		148	141	142	269	187	127	116	120	154	127	90	**89.5-269.1**	**147.4**
PCB, ng/SPMDxday^{-1}		3.5	3.2	3.8	2.8	2.1	2.5	2.5	3.7	2.1	1.8	2.1	**1.8-3.8**	**2.7**
HCB, ng/SPMDxday^{-1}		2.1	1.9	3.1	2.2	1.2	1.3	1.5	2.1	1.6	0.7	1.2	**0.7-3.1**	**1.7**

*Daily limit value, $\mu g/m^3$, Lithuanian hygiene norm HN 35:2007.
** Sum for all xylene

3.1.2. Nitrogen dioxide

The concentrations of NO_2 ranged from 1.0 μg/m^3 (B10) to 5.4 μg/m^3 (B11), with the median of 4.0 μg/m^3. Such values were generally lower compared to our previous measurements in multifamily apartments (median 13.1 μg/m^3) (Du et al., 2015) as well as studies in other areas, such as Sweden (10.1 μg/m^3 in passive buildings) (Langer et al., 2015) and Denmark (below 10 μg/m^3) (Sorensen et al., 2005). The investigated buildings in Lithuania did not have indoor combustion sources and were located further from outdoor traffic sources, thus resulting in low concentrations of indoor NO_2.

3.1.3. Formaldehyde

The concentration of the only measured VVOC, formaldehyde, ranged from 3.3 μg/m^3 (B10) to 52.3 μg/m^3 (B7) with overall measured median value of 30.8 μg/m^3 (**Table 3.1**). Thus the concentration of formaldehyde exceeded the Lithuanian national standard limit daily value of 10 μg/m^3 in all buildings, except the unoccupied B10 (HN 35:2002). It should be noted that the Lithuanian standard limit daily value is very low. WHO guideline exposure threshold values are 100 μg/m^3 (WHO guidelines for IAQ, 2010). Unfortunately, WHO defines the same value for both short term as well as long term exposure (WHO guidelines for IAQ, 2010). The concentrations of formaldehyde in 10-year-old B8 was also quite high (37.5 μg/m^3). Such trend of increased levels of formaldehyde has been noticed during our earlier studies. In multifamily apartment settings the median formaldehyde concentration was 21.2 μg/m^3 (Du et al., 2015). At the same time, formaldehyde has been reported as an issue in multiple earlier studies, i.e. average formaldehyde concentrations in newly built French buildings were 45 μg/m^3, while in 1-3-year-old buildings 35 μg/m^3 (Marchand et al, 2008). The median concentration of formaldehyde in new energy efficient buildings in France was lower (20.5 μg/m^3) (Derbez M, et al., 2014a) or (20.8-27.8 μg/m^3) (Derbez et al., 2014) than in this study. Langer et al., (2015), also reported lower median concentrations of formaldehyde in passive, conventional new buildings and the older single-family buildings in Sweden (11, 16 and 22 μg/m^3 respectively) (Langer et al., 2015 and Langer and Beko, 2013). Similar to this study formaldehyde concentrations ranging from 25.7 to 32.8 μg/m^3 were reported in two zero-energy homes in Massachusetts at medium ventilation flow (Engelmann et al., 2015). Formaldehyde is used in the production of phenolics, urea, melamine, and polyacetal resins for adhesives and binders in wooden products, paper, and synthetic fibre products. It is also directly used in aqueous solutions (like formalin) as a disinfectant and preservative. Such applications make wooden products, carpets, paints, and varnish the main indoor sources of formaldehyde (Kolarik et al., 2012). Formaldehyde concentrations do not simply result from additive emissions from the materials (Gunschera et al., 2013). This data indicates that low energy buildings in many countries do not correspond to "green" or "sustainable" buildings with respect to the materials used for surfaces or furniture. Although the low energy building concept is primarily energy-oriented, the issue of

extending the conscious approach in building/buying low energy buildings must be supplemented by further knowledge of installing environment friendly low emitting interior materials.

3.1.4. VOCs

The investigated sample of 11 buildings reflected two patterns of IAQ based on concentrations of construction related organic compounds. The first group included the majority of buildings which were built at least one year ago and had low emissions from surfaces. Another group was represented by several buildings which had recently completed building works and subsequently high emissions from one or several sources.

The first round of investigations

The first group included B1-B3, B6-B9, and B11. Total BTEX concentration among these buildings ranged from 5.0 µg/m^3 (B6) to 10.0 µg/m^3 (B1) (**Table 3.1**), with the median value of 6.1 µg/m^3. Although within this study these emissions were relatively low, they were higher than the previously measured BTEX average summer time concentrations of 3.1 µg/m^3 in ordinary single-family buildings in Kaunas (Kliucininkas et al., 2014). On the other hand, such concentrations were lower compared to BTEX levels in apartments in multi-family buildings (median 13.53 µg/m^3) but higher compared to Finnish apartments (5.68 µg/m^3) (Du et al., 2015). It should be noted that sampling in the latter study was performed during the cold season and may have had different sources of BTEXs indoors.

The above provided data reported results from the set of buildings having rather uniform and low concentrations of pollutants. Another group of recently established buildings revealed elevated VOC concentrations. In B4 and B5, elevated concentrations of BTEX, styrene, trichloroethylene, and formaldehyde have been registered. For example, in B4 p-xylene concentration reached 214.5 µg/m^3, o-xylene – 328.9 µg/m^3, ethylbenzene – 521.7 µg/m^3, and styrene – 419.0 µg/m^3, thus exceeding Lithuanian standard limit daily values for indoor air. This sample might have been affected by the air from a recently painted garage using a solvent based paint. Elevated levels of VOCs, especially toluene, trichloroethylene and formaldehyde in the unoccupied B5 were caused by some recently installed or painted interior surfaces using latex based paint. In B10 where no interior installations had been done VOC concentrations were remarkably lower. We do not consider high concentrations as outliers due to experimental error. Such phenomenon of high indoor VOC concentrations has been reported by other authors. Liang et al., (2014), investigated VOC concentrations in a new apartment in Beijing from the beginning of interior construction to completion as a case study. A total of five construction stages, namely the putty, wall paint, door and doorframe, cupboard and furniture had been evaluated. Concentrations as high as 664 µg/m^3 for toluene were detected after installation of the wooden door and doorframe, and 255 µg/m^3 for benzene, 162 µg/m^3 for ethylbenzene, 107 µg/m^3 for p-xylene, 185 µg/m^3 for styrene and 85.1 µg/m^3 for o–xylene after furniture had been installed (Liang et al.,

2014). In our case, we attribute these elevated concentrations to covering of wall surfaces with latex-based paint as well as the application of solvent-based varnish on wooden interior surfaces. The reduction of these concentrations in time is further described in the second round of investigation.

The median concentrations of benzene, toluene, and xylenes was very close to those reported in Swedish passive and conventional new buildings (Langer et al., 2015), but concentrations of individual BTEX compounds and styrene where slightly lower compared to those measured in newly-built energy efficient buildings in France (Derbez et al., 2014a). The median concentrations of benzene, toluene and xylenes in this study was 3–7 times lower than in two zero–energy homes in Massachusetts at medium ventilation flows (Engelmann et al., 2013). In contrast to this study trichloroethylene and tetrachloroethylene were not detected in most of the French buildings (Derbez et al., 2014a). As expected, the relationships between VOC concentrations were strong, indicating similar emission sources (**Table 3.2**). The concentration of formaldehyde was not correlated with the concentrations of the rest of the VOCs, but was correlated with NO_2 concentration (r=0.71). We attribute this correlation more to the accidental relationship than a proof of the same emission sources. Wood based panels/products typically emit formaldehyde. It may also originate from unvented fuel burning appliances or outdoor pollution from traffic. The indoor formaldehyde concentrations are usually several times higher than the outdoor ones (Marchand et al., 2008). On the contrary, the concentration of NO_2 is usually higher outdoors because it primarily originates from combustion processes (street traffic), but also during indoor combustion activities. Langer et al., (2015), reported the average I/O ratio for NO_2 in spring-summer as 1.8 for the passive and conventional buildings, suggesting some indoor sources (Langer et al., 2015). In our case, indoor fuel combustion was not present; outdoor combustion sources were not at a close proximity, thus a definite answer on the relationships cannot be provided.

Table 3.3. Spearman correlation coefficient (p<0.05) matrix of the concentrations measured.

	CO_2	NO_2	Benzene	Ethyl-benzene	Toluene	o-Xylene	p-Xylene	BTEX	Styrene	TCE	PCE	Formaldehyde	Sum PAHs	Sum alkyl-PAHs	PCBs
Benzene	0.15	0.25													
Ethylbenzene	0.21	0.25	0.81												
Toluene	-0.01	0.44	0.87	0.77											
o-Xylene	0.17	0.45	0.78	0.59	0.81										
p-Xylene	0.01	0.29	0.76	0.49	0.7	0.68									
BTEX	0.13	0.31	0.92	0.77	0.68	0.91									
Styrene	0.09	0.36	0.4	0.46	0.67	0.69	0.76								
Trichlorethylene (TCE)	0.06	0.24	0.76	0.76	0.77	0.59	0.49	0.77	0.77						
Tetrachlorethylene (PCE)	0.15	0.32	0.67	0.81	0.61	0.81	0.7	0.68	0.67	0.69					
Formaldehyde	0.5	0.71	0.32	0.26	0.38	0.13	0.27	0.25	0.36	0.38	0.16				
Sum PAHs	0.18	-0.01	0.37	0.7	0.56	0.51	0.17	0.4	0.11	0.3	0.4	0.09			
Sum alkyl-PAHs	0.1	-0.31	0.31	0.35	0.17	0.38	0.05	0.39	0.08	0.26	0.44	-0.39	0.61		
PCBs	0.22	0.39	0.38	0.45	0.48	0.51	0.11	0.28	0.12	0.22	0.36	0.22	0.44	-0.05	
HCB	0.18	0.34	0.55	0.57	0.48	0.62	0.13	0.48	0.2	0.34	0.59	0.06	0.44	0.23	0.89

The second round of investigations

The results of VOC concentration measurements in B5 during five consecutive weeks after the complete installation of the interior are presented in **Table 3.3**. The latest interior works in the building were the installation of wood based floor panel boards and solvent based varnishing of wooden stairs. The experiment started on the third day after completion. Extremely high concentrations of toluene, xylenes and ethylbenzene were detected initially. Concentrations of all VOCs investigated clearly had a decreasing tendency with the exception of trichloroethylene, its concentration was increasing. Trichloroethylene (TCE) is used mainly as a solvent to remove grease from metal parts, but it is also an ingredient in adhesives, paint and spot removers (ASTDR). The highest emission factors of TCE were reported from OSB boards (1.4 $\mu g/m^3/h$), followed by solid wood (0.42 $\mu g/m^3/h$) and plywood (0.28 $\mu g/m^3/h$) (National Research Council of Canada). The increase of TCE concentration in time could be presumed as being influenced by chemical reactions, e.g. TCE is a sub-product of PCE degradation.

Table 3.4. VOC concentrations ($\mu g/m^3$) estimated during five consecutive weeks after the installation of the interior.

Week	Benzene	Ethylbenzene	Toluene	o-Xylene	p-Xylene	BTEX	Styrene	TCE	PCE
1	16.6	126.3	812.4	100.9	188.8	1366.4	60.6	4.9	5.2
2	3.1	75.5	377.5	66.4	113.4	753.7	58.9	7.0	3.9
3	24.4	32.1	113.3	35.3	53.0	295.0	18.4	9.1	1.0
4	10.2	14.7	19.0	10.6	24.9	82.3	1.5	8.5	1.2
5	7.2	2.5	13.1	2.3	4.2	31.8	1.2	17.1	2.0

The decrease of VOCs had exponential pattern ($y = 4{,}438.4e^{-0.975x}$, $R^2 = 0.988$). Already during the fourth week the concentrations of VOCs reached the acceptable levels below limit values. The decrease ranged from 38.9% (benzene) to 97.7% (toluene). Presumably, high initial VOC concentrations were determined by quickly emitting VOC emission sources like paints or varnishes. The dominating compound toluene is known to be one of the fast dissipating VOCs (Shin and Jo, 2012). Jarnstrom et al., (2006), also reported a rapid decrease of TVOC concentrations in a new building in Finland. The indoor air TVOC concentrations in the occupied apartments decreased 60-75% to a concentration level of 300-500 $\mu g/m^3$ during 6 weeks (Jarnstrom et al., 2006).

3.1.5. SVOCs (PAHs, PCBs, and HCB)

The PAHs were rather uniformly distributed among the buildings, ranging between 29.7 and 94.3 ng/SPMD/d (**Table 3.1**). This implies that there were no

major sources of PAHs neither indoors nor outdoors of the tested buildings. The highest levels were observed of phenanthrene (441-1270 ng/SPMD/d), fluorene (59-651 ng/SPMD/d), naphthalene (53-409 ng/SPMD/d) and fluoranthene (56-180 ng/SPMD/d). The mean across all measured buildings of sum 16 EPA PAHs in SPMDs was 53.5 ng/SPMD/d, which is higher than estimated previously in Kaunas, when sum PAH amount ranged 14-19 ng/SPMD/d (Kauneliene et al., 2015). Strandberg et al., (2006), reported similar PAH levels 30-60 ng/SPMD/d in the air of Swedish buildings with no biomass burning while in buildings with biomass burning considerably higher PAH levels (30-350 ng/SPMD/d) were reported (Strandberg et al., 2006). Similar mean PAH level (37.8 ng/SPMD/d) were estimated inside 52 homes situated along the border between Arizona and Mexico (Gale et al., 2009).

The amounts of alkylated PAHs ranging from 89.5 to 269.1 ng/SPMD/d were considerably higher than the amount of un-substituted PAHs. The highest amounts were observed of trimethylnaphthalene (3.5-120.4 ng/SPMD/d), dimethylnapthalene (5.4-67.5 ng/SPMD/d), methylnapthalene (2.2-26 ng/SPMD/d) and dimethylphenanthrene (1-26.5 ng/SPMD/d). In Swedish buildings with no wood burning methylated phenanthrenes were dominating among the alkylated species but at lower levels, not exceeding 9.1 ng/SPMD/d (Strandberg et al., 2006). In the USA homes about 60 alkylated PAHs were detected. The total estimated amounts of alkylated PAHs in the USA homes ranged from 3 to 33 ng/SPMD/d (Gale et al., 2009). The scientific evidence of alkylated PAH origin, emissions and fate is very scarce.

The highest amounts of PAHs and alkylated PAHs were detected in B4 and B5 also having the highest concentrations of VOCs. This suggests similar emission sources. The amounts of PAHs were strongly correlated (r_s=0.70) with the concentration of ethylbenzene, a weaker correlation was observed in case of toluene (r_s=0.56) and o-xylene (r_s=0.51). It is well documented that the major indoor sources of PAHs include outdoor air, cooking, smoking and unvented combustion appliances (Weschler 2011), while the indoor sources of VOCs include household products, fragranced consumer products, off-gassing of building materials, floor and wall coverings, paints and adhesives, deodorisers, fuel evaporation, tap water and bleach use and some combustion processes (e.g. cooking, smoking) (Bari et al., 2015). Presumably, PAHs as well as alkylated PAHs were emitted from building materials, household products or petroleum products in case of air exchange with the garage. Glader et al. investigated patterns of organic compound emissions in building structures and did report the existence of PAHs in building materials and the fact that PAHs were more strongly associated with structures in which the predominant construction material was concrete, than in structures mainly made of wood (Glader and Liljelind 2012).

The concentrations of PCBs ranged from 1.8 to 3.8 ng/SPMD/d and were dominated by DiCB and TriCBs. These values are comparatively low. Similar amounts (<0.01-2.8 ng/SPMD/d) were reported in Sweden (Strandberg et al., 2006), and slightly lower (1.1-13 ng/SPMD/d) than reported in USA buildings (Gale et al., 2009). Amounts of hexaclorbenzene (HCB, 0.7-3.1 ng/SPMD/d) in this study were

lower than in the USA buildings, 8-220 ng/SPMD/d (Gale et al., 2009). PCB sources indoors include stabilisers for PVC wire insulation, flame-retardants, pesticide extenders and additives (in sealants, adhesives, paints, and floor finishes) (Weschler 2011). Due to adverse environmental and health effects, it was banned in many countries and included in the Stockholm Convention (Stockholm Convention, 2011), which aims to protect the environment and human health from persistent organic pollutants. Low concentrations of these pollutants in the currently investigated buildings confirm the effectiveness of the international legislation, reducing exposure of residents.

3.2. Characterisation of active indoor pollution sources

3.2.1. PM emissions from tested pollution sources

The performed measurements allowed the generation of a database of the temporal variation profiles of various indoor air pollution sources (20 different sources) as measured by several aerosol devices. **Table 3.4** presents the measured maximum PNCs in the $PM_{0.01-0.3}$ and $PM_{0.3-10}$ ranges (ACR 0.5 ± 0.1 h^{-1}). As well as the change in T and RH values during each operation. This section discusses the data which was obtained from the in-duct measurements.

The highest concentration of $PM_{0.01-0.3}$ was observed during the operations of thermal sources, including the hair dryer operating at maximum setting (4.28×10^6 p/cm^3 with a mode at 21 nm), heating an empty Teflon pan (2.58×10^6 p/cm^3, 37 nm), and onion frying in a Teflon pan (1.17×10^6 p/cm^3, 49 nm), as well as a cigarette smouldering (5.43×10^6 p/cm^3, 27 nm). In this study the hair dryer operating at maximum setting generated more ultrafine particles (UFP, in our case represented by $PM_{0.01-0.3}$) than reported by Glytsos et al., (2010) (5.7×10^5 p/cm^3). The Teflon pan was confirmed to be a strong source of UFPs (Afshari et al., 2005) as well.

As expected, cooking related sources generated large amounts of UFP. The maximum measured $PM_{0.01-0.3}$ concentration varied from 3.48×10^4 (electric stove operating alone) to 2.58×10^6 p/cm^3 (Teflon pan heating on the electric stove), except for the water boiling on the electric stove, 3.48×10^3 p/cm^3. Findings are comparable to the data reported in the study of Hussein et al., (2006), where the emission of PM from cooking activities reached 1.8×10^6 p/cm^3 (more than 90% of the total amount was UFP). Previously performed studies revealed that maximum total PNC of particles may vary between 1.5×10^6 and 5.6×10^6 p/cm^3 during various cooking activities (Afshari et al., 2005; Dennekamp et al., 2001; Morawska et al., 2003 and He et al., 2004).

The maximum observed $PM_{0.01-0.3}$ number concentration ($PNC_{0.01-0.3}$) resulting from unscented and aromatic paraffin candle burning were almost equal (3.98×10^5 and 3.92×10^5 p/cm^3, respectively) and unimodal, with a slight difference in modes 15 and 12 nm. Glytsos et al., (2010) previously reported that PNC varied in the range of $2.97\times10^5\pm1.94\times10^5$ p/cm^3, which is comparable to the results obtained in this study. Pagels et al., (2009) previously reported the total measured PNC (16-1000 nm) varied in the range from 2.7×10^5 to 11.4×10^5 p/cm^3.

In the fine/coarse range ($PM_{0.3-10}$, measured with OPC), cooking oil heating revealed the highest $PM_{0.3-10}$ number concentration ($PNC_{0.3-10}$) of 1.82×10^3 p/cm^3. Relatively high concentrations were registered during cigarette (1.25×10^3 p/cm^3) and incense stick (1.02×10^3 p/cm^3) smouldering. Some of the sources did not show significant $PM_{0.3-10}$ emission, including the electric air heater, electric stove, and floor wet mopping with detergent, and interestingly, floor vacuuming. Low emissions of $PM_{0.3-10}$ from floor vacuuming was reported earlier, e.g., Afshari et al., (2005), where a vacuum cleaner with a full dust bag generated particles in the size ranges of 0.4-0.5 and 0.5-0.6 µm (12 and 4 p/cm^3, respectively) and UFP 2.1×10^4 p/cm^3. Similarly, to our study, a modern vacuum cleaner equipped with a HEPA filter was used, which does not emit a substantial amounts of coarse particles.

Table 3.5. Summary of maximum PNC from 20 tested indoor activities, sampling in the exhaust channel and in the centre of the ceiling. The standard deviation (SD) represents variation between mean values of repeated experiments.

Indoor air pollution sources	Sampling in the exhaust channel		Sampling in the centre of the ceiling		Change in T ± SD	Change in RH ± SD
	$PM_{0.01-0.3}$ C_{max} ± SD×10^3, p/cm^3	$PM_{0.3-10}$ C_{max} ± SD, p/cm^3	$PM_{0.01-0.3}$ C_{max} ± SD×10^3, p/cm^3	$PM_{0.3-10}$ C_{max} ± SD, p/cm^3	°C	%
Cooking oil heating on electric stove	1,555±279	1,824±506	2,215±327	1,097±168	3.3±0.25	1.1±0.08
Electric stove operating alone	35±7	3±2	39±5	4±2	3.9±0.31	1.9±0.34
Onion frying in oil in a Teflon pan on electric stove	1,175±144	649±362	1,111±147	355±144	2.5±0.22	14.6±2.04
Teflon pan heating on electric stove	2,583±328	576±145	3,548±363	857±269	3.2±0.30	1.4±0.12
Water boiling on electric stove	411±49	172±42	573±67	66±26	3.1±0.28	42.1±3.79
Aromatic candle burning	392±47	23±7	231±35	64±13	1.4±0.11	0.6±0.05
Cigarette burning	543±113.4	1,253±421	668±88	664±168	0.9±0.04	1.9±0.11
Incense smouldering	63±11	1,015±150	51±8	1,227±178	0.6±0.03	1.6±0.15
Paraffin candle burning	399±32	25±8	508±7	85±16	1.4±0.09	0.3±0.02
Hair dryer operating (hottest air)	4,277±589	13±5	4,011±451	10±3	2.1±0.24	0.5±0.06
Hair dryer operating (medium hot air)	863±135	6±3	396±58	5±1	1.5±0.14	0.4±0.04
Hair spray spraying	7±1	378±82	2±0.4	159±53	0.9±0.03	0.2±0.02
Air-freshener (aerosol) spraying	1±0.1	31±14	4±0.4	12±2	0.7±0.02	1.8±0.15
Electric air heater operating	252±44	2±1	485±86	2±1	7.7±1.04	2.2±0.25
Electric radiator operating	4±0.6	4±1	1±0.2	4±2	3.3±0.41	2.2±0.28
Floor vacuuming	1.5±0.1	2±1	5±0.6	2±1	3.6±0.34	0.7±0.06
Floor wet mopping with detergent	17±3	3±2	5±0.8	3±2	0.8±0.06	14.6±2.12
Furniture polisher spraying	6±0.6	70±16	2±0.2	90±26	0.6±0.03	0.8±0.04
Ironing with steam	3±0.58	61±12	36±6	245±36	1.5±0.13	13.4±1.14
Ironing without steam	19±2.4	6±2	28±4	36±17	1.4±0.09	1.1±0.07

3.2.2. Assessment of temporal variation patterns of pollution sources

The variation of pollution sources in time and subsequent variation of IAQ are important, since they may provide background data for air quality control algorithms. So far, the management of residential IAQ was mostly based on the real-time measurements of thermal comfort, and CO_2, as the indicator of human activity. Our data provides information that may be utilised considering measurements of aerosol particles as indicators of IAQ. Depending on the rates of temporal variation of pollutant concentrations resulting from the active indoor sources, the IAQ management systems may assume different algorithms for achieving suitable IAQ.

The pollution episodes generated in the chamber were fitted to increase-decrease patterns, as described in the Methods section. An example of the temporal variation of PNC during hair spray (5 s) and onion frying operations (10 min) are presented in **Figure 3.1**. The maximum $PNC_{0.01-0.3}$ was observed after 12 min following the hair spray and 7 min after onion frying in a Teflon pan. In both of these cases, the responses from PM monitors were rapid and significant. At the same time, the concentration was fluctuating substantially during both increase and decay phases, possibly reflecting the turbulence of air flow within the chamber. A similar fluctuation has been registered in case of most pollution sources. This may pose some challenges in real-time prediction of increase and decay rates, since management algorithms may anticipate a faster or slower or even false increase or decrease in PM concentrations. This can be solved by the application of time series smoothing algorithms, which would even out the fluctuations during a given time period. Here we assume that the management algorithm would be based on the increase or decrease rate of concentrations, not just by a single signal of reaching threshold value, which is often the case with T or CO_2 based management of indoor environmental quality.
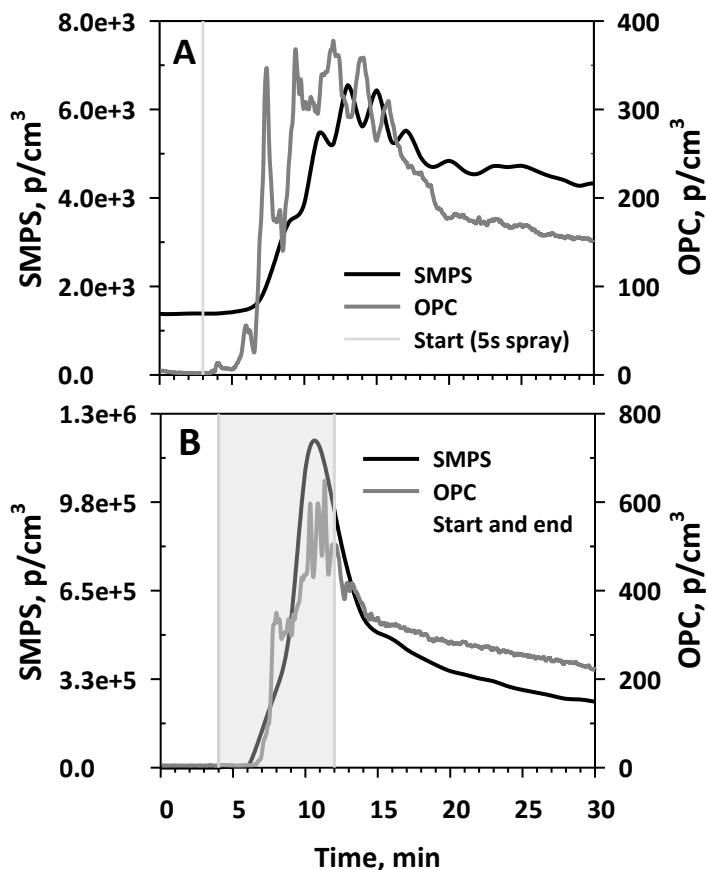
Figure 3.1. Responses and dynamics of particle concentrations in the ventilation exhaust in case of hair spray (A) and onion frying in a Teflon pan operations (B).

The analysis of the variation of particle size distributions in time has revealed the expected transformation processes in the aerosol size, such as particle nucleation, growth, and coagulation. The nucleation and growth were observed in case of sources emitting VOCs which served as a precursor of UFP formation (**Figure 3.2. A**, hair spray). The formation of UFP occurred 10-15 min from the start of the operation. Cooking operations (such as onion frying, **Figure 3.2. B**) revealed that most of the particle emission occurred in the UFP range, which agrees with the findings by Afshari et al., (2005) and Glytsos et al., (2010). The dynamics of PSDs from another 18 tested indoor activities are provided in the supporting information (**ANNEX 1.**). Generally, most indoor pollution sources were associated with elevated indoor $PM_{0.01-0.3}$ concentrations, which remained elevated during the period of 30 min at 0.5 h^{-1} ACH. It has been shown that elevated concentrations can remain for up to 12 hours (Wierzbicka et al., 2015) at low air change rates. This indicated that a standard ventilation rate may not be sufficient to improve IAQ based on UFP concentrations and further modelling should be carried out to adjust the air change rate to effectively reduce the concentrations.
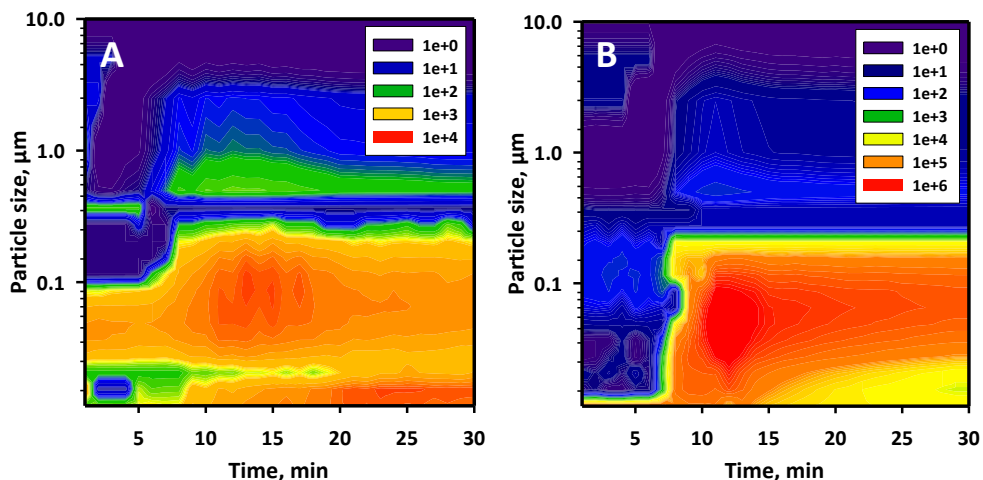
Figure 3.2. Dynamics of PSD during hair spray (A) and onion frying in a Teflon pan (B) operations (dN/dlogDp, p/cm^3).

Increase and decay rates for $PNC_{0.01-0.3}$ and $PNC_{0.3-10}$ are presented in **Table 3.5**. These rates were obtained from in-duct measurements. Based on these rates, the tested sources were grouped to rapid processes (the maximum UFP was measured after 1-2 min after the pollution episode was generated) and slow processes (7-25 min). In accordance with the slope coefficient *b* (representing the rate of increase in concentrations) of the sigmoidal function, the rapid sources were classified as having *b*≤30, and slow as *b*≥30. The fast processes included mostly thermal sources ("other thermal sources", electric stove operating alone in "cooking related sources", electric air heater, and ironing without steam), as well as air-freshener and hair dryer (the hottest air setting) from "personal care sources". Most operations emitting PM precursors (hair spray, furniture polisher, air-freshener spraying, floor vacuuming, floor wet mopping with detergent, etc.) may be attributed to slow processes, since the maximum PM concentration was reached only after 20-25 min from the start of the experiment.

The evaluation of d_1 (fast decay phase) and d_2 (slow) parameters of the exponential decay function (**Table 3.5**) revealed that $PNC_{0.01-0.3}$ decay rate depends on the duration of the pollution source. Longer lasting sources emit higher concentrations of PM, requiring more time for the dispersion and removal, thus $PNC_{0.01-0.3}$ produced "flatter" decay curves (e.g. aromatic and paraffin candle burning, cooking oil heating, onion frying, etc.).

In some cases, corresponding to the UFP precursor emission or overall low PM emissions, the decay of particle concentration could not be adequately represented in a 30 min period due to continuously elevated concentrations (floor wet mopping with detergent, floor vacuuming, electric radiator operating alone, ironing with steam, and water boiling).

The above presented data provide several indications with respect to using it as a control input for IAQ management. Most of the sources had well-established

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dynamic patterns, with a clear increase and decay rate, which may serve as indicators of the process rate and be used for controlling of IAQ management systems. At the same time, due to turbulence inside the chamber and non-uniform emission of pollutants from the source, the concentration was fluctuating during the increase, and this potentially may require some time series smoothing algorithms to be applied in order to obtain a representative increase rate of the pollutant concentration in the air. The results provided here do not cover the generation of pollution from mixed sources, which may cause even more complicated increase/decrease patterns.

At the same time, UFP are of specific health concern, the complex formation and transformation mechanisms make them different from other pollutants, which have more predictable increase/decrease patterns, related to source activity. A slow decay rate of $PM_{0.01-0.3}$ concentrations in air suggest that they are a very important parameter for IAQ control, since the management based on the UFP concentration may require higher and prolonged ventilation or cleaning rates.

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Table 3.6. Summary of $PNC_{0.01-0.3}$, $PNC_{0.3-10}$ slope coefficients, time to reach max concentrations and decay rates from the tested indoor activities (SD – standard deviation).

Indoor air pollution sources	$PNC_{0.01-0.3}$ slope coefficient $b \pm$ SD	Time to reach max $PNC_{0.01-0.3}$ concentrations ± SD, min	$PNC_{0.01-0.3}$ decay rate $d_1 \pm$ SD, h^{-1}	$PNC_{0.01-0.3}$ decay rate $d_2 \pm$ SD, h^{-1}	$PNC_{0.01-0.3}$ slope coefficient $b \pm$ SD	Time to reach max $PNC_{0.01-0.3}$ concentrations ± SD, min	$PNC_{0.01-0.3}$ decay rate $d_1 \pm$ SD, h^{-1}	$PNC_{0.01-0.3}$ decay rate $d_2 \pm$ SD, h^{-1}
Cooking oil heating on electric stove	44.6±9.7	8±1	27.7±4.8	1.8±0.3	27.3±5.2	12±2	7.4±1.4	0.1±0.01
Electric stove operating alone	9.1±1.9	3±1	25.2±4.5	21.2±7.8	-	-	-	-
Onion frying in oil in a Teflon pan on electric stove	46.0±14.5	7±1	29.9±4.8	2.2±0.5	51.8±8.7	8±1	7.8±2.5	0.3±0.02
Teflon pan heating on electric stove	40.5±7.3	8±1	26.6±5.6	-	52.5±9.3	10±1	6.3±2.3	1.1±0.1
Water boiling on electric stove	282.1±39.5	10±2	-	-	91.2±17	11±2	11.2±1.4	0.7±0.1
Aromatic candle burning	8.5±2.6	2±1	63.8±16.2	3.6±0.6	1.7±0.2	1±0	12.4±1.9	0.8±0.1
Cigarette burning	10.6±1.6	2±1	74.9±13.2	3.6±0.9	1.3±0.1	1±0	2.5±0.7	2.4E-11±1.1E-11
Incense smouldering	3.1±0.5	2±1	11.2±2.6	0.4±0.1	1.4±0.1	1±0	3.0±0.6	0.1±0.01
Paraffin candle burning	11.4±3.1	2±1	60.5±14.2	0.1±0.01	4.5±1.5	1±0	13.2±2.8	7.0E-09±2.1E-09
Hair dryer operating (hottest air)	5.3±2.3	2±1	24.5±3.6	2.2±0.9	-	-	-	-
Hair dryer operating (medium hot air)	32.9±13.6	5±1	4.3±0.8	3.6±1.1	-	-	-	-
Hair spray spraying	206.2±25.6	15±2	15.5±5.1	0.4±0.1	59.6±9.2	8±1	2.0±0.4	1.1E-10±1.2E-12
Air-freshener (aerosol) spraying	4.0±1.3	2±1	85.7±18.9	7.2±2.3	99.2±15	4±2	1.4±0.1	1.2E-10±1.2E-11
Electric air heater operating	0.8±0.2	8±2	34.9±8.3	1.1±0.2	-	-	-	-
Electric radiator operating	330.9±72.9	15±2	-	-	-	-	-	-
Floor vacuuming	394.2±86.3	10±2	-	-	-	-	-	-
Floor wet mopping with detergent	111.5±21.4	25±5	-	-	-	-	-	-
Furniture polisher spraying	161.0±23.3	2±1	78.8±16.4	0.7±0.1	112.4±2		1.7±0.2	0.1±0.01
Ironing with steam	79.9±17.8	8±1	-	-	40.5±4.6	4±1	1.3±0.3	1.6E-09±1.5E-10
Ironing without steam	18.6±5.6	5	171.4±17	0.7±0.1	-	-	-	-

3.2.3. Comparison of monitoring in the exhaust air duct vs. centre of a ceiling

Two different measurement locations, the exhaust air duct and centre of the ceiling were tested as potential sites for placing IAQ monitoring sensors. The measurement of pollutants in the exhaust air duct of the forced ventilation system provides a convenience for concentrating the air from the entire room into a single flow. Moreover, samplers operating without pumps may benefit from already existing flow, thus eliminating the need of sampler pumps and subsequent service or noise burden. At the same time, the centre of the ceiling has been long utilised for placing fire detectors as the most representative location, thus it was of interest to compare variation of pollutants between the two locations. The CFD confirmed that the generated pollution plumes affect both measurement sites (**Figure 3.3 and ANNEX 2**), but the experimental study was used to research quantitative differences.
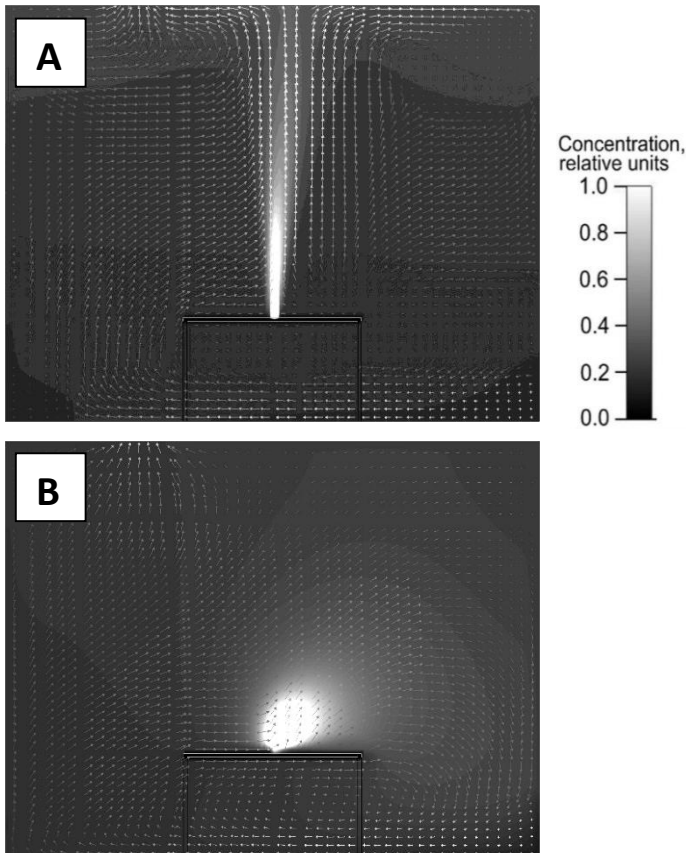
Figure 3.3. CFD prediction results of air velocity vectors and $PM_{0.5}$ particle concentration contours in the case of warm contaminant injection (A) and isothermal contaminant injection (B).

The temporal variation of $PNC_{0.01-0.3}$ during cigarette (1 min of continuous burning) and paraffin candle burning (10 min of continuous burning) in the two sampling locations is presented in **Figure 3.4**. The maximum $PNC_{0.01-0.3}$ in the cigarette burning experiment were 5.43×10^5 p/cm^3 in the exhaust channel (which corresponds to the data presented in Chapter 3.1) and 6.68×10^5 p/cm^3 in the centre of the ceiling. The difference between maximum concentrations (ΔC) was found to be statistically insignificant ($p>0.05$) based on the Mann–Whitney U test. The same applies to the candle burning experiment. Statistically significant differences ($p<0.05$) in maximum concentrations were observed in cases of highly emitting sources, such as Teflon pan heating, cooking oil heating, and onion frying, while all remaining pollution sources revealed no statistically significant difference between two measurement locations.
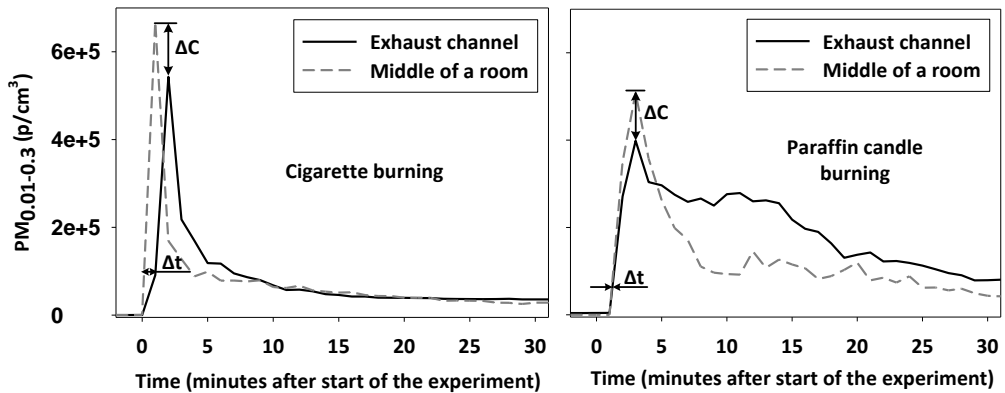
Figure 3.4. Difference in detection time and particle concentration between two sampling locations (exhaust channel and centre of the ceiling) for cigarette and paraffin candle burning, (ΔC – concentration difference between maximum PM levels, Δt – difference between sensor response time).

The difference in the detection time (Δt) between the two locations was rather low in the case of both exhaust ventilation and the centre of the ceiling locations. Both locations responded within two minutes from the initiation of the source. The one-minute temporal resolution did not allow a finer discrimination between the detection times. It may be argued, whether this resolution should be increased for long-term continuous monitoring. It may be so that the position of the pollution source and the exhaust diffuser within a room plays a higher role in the response of the sampler.

The twenty tested sources provided a sufficient sample to run a regression analysis to establish a relationship between these two measurement locations (**Figure 3.5**). The relationship between the detection time in the exhaust channel and in the centre of the ceiling is presented in **Figure 3.5 (A)**. The sources displayed the dynamic profiles similar to those reported in the Chapter 3.2.1. The response time ranged from less than a minute for combustion processes – cigarette, incense, paraffin and aromatic candle, up to several tens of minutes (e.g. floor moping with a detergent), indicating formation of aerosol from gaseous precursors. The regression

model has represented 78% of the data variation. The detection times of $PNC_{0.01-0.3}$ in the centre of the ceiling were slightly faster in comparison with exhaust air duct measurements, as indicated by the slope, equal to 0.88. This may be associated with the particular positioning of the pollution source within a room; consequently, pollutants were not efficiently transported to the upper layer of the chamber. This representation may be different for other locations of the inlet and outlet diffusers, room dimensions, and air change rate. On the other hand, the obtained good agreement shows that both sampling locations may work well for the representation of IAQ dynamics within the room.
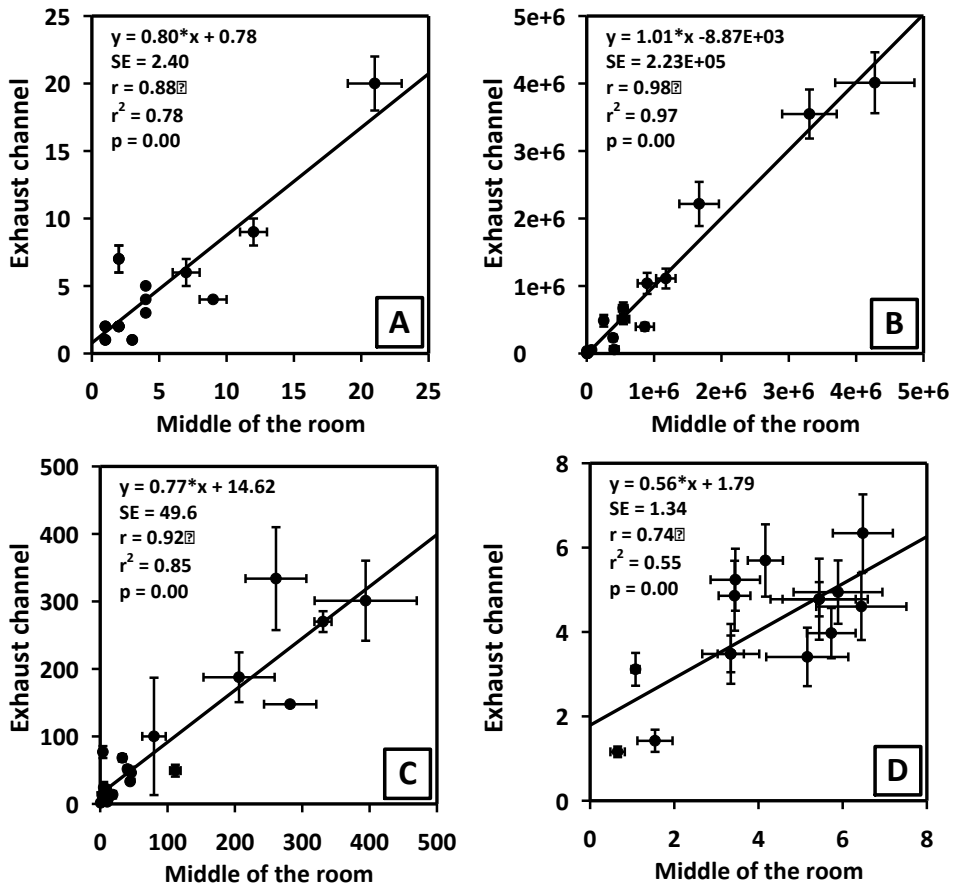
Figure 3.5. The correlation between the $PM_{0.01-0.3}$ detection time (A, min), maximum assessed $PNC_{0.01-0.3}$ (B, #/cm^3), slope at the inflection point (C), and $PM_{0.01-0.3}$ concentration decay rate (D, h^{-1}) in different sampling location: in the exhaust channel and in the centre of the ceiling.

Figure 3.5 (B) represents the relationship between the maximum $PNC_{0.01-0.3}$ in both measurement locations. The agreement between measurement locations was high ($R^2=0.98$). Both measurement locations represented high-emitting and low-emitting sources, with the regression slope almost equal to unity.

Figure 3.5 (C) shows the relationship between the $PNC_{0.01-0.3}$ increase rate between the initial and the maximum observed values (parameter *b* from sigmoidal equation). The agreement was linear and strong ($R^2=0.85$). The agreement for the decay rate, as seen in **Figure 3.5 (D)**, was lower ($R^2=0.55$), but still sufficient to establish a statistically significant relationship ($p<0.05$). The representation of increase and decay rates of $PNC_{0.01-0.3}$ was better during the measurements in the centre of the ceiling, similarly with the response time. Again, the positioning of the source in the room and the air change rate may have had the governing effect, but this may represent rooms in a majority of dwellings, unless the pollution source is recessed or obstructed by irregular walls or furniture.

3.2.4. VOCs emissions from tested pollution sources

The dynamics of pollution concentrations under different pollution sources were diverse throughout the 30 min monitoring period, maximum concentrations were reached at different points of time. Most of the concentration increase patterns can be described by the empirical first order exponential growth equations the can be further applied to the forecasting and real-time control of the IAQ.

The tested pollution sources revealed a broad range of pollutant emission patterns, including responses of tested VOC sensors. Among the gaseous pollutant emissions, the hair spray has impacted most of the gaseous pollutant (CO and VOC) concentrations. The maximum measured CO concentration was equal to 14 ppm and VOCs to 19.16 ppm above the background level.

Figure 3.6 shows the dynamics and concentration levels of cigarette smouldering as an example of several researched cases. In both of these cases, the responses from both gaseous monitors were rapid and significant, showing correlation between the readings of different measurement techniques.
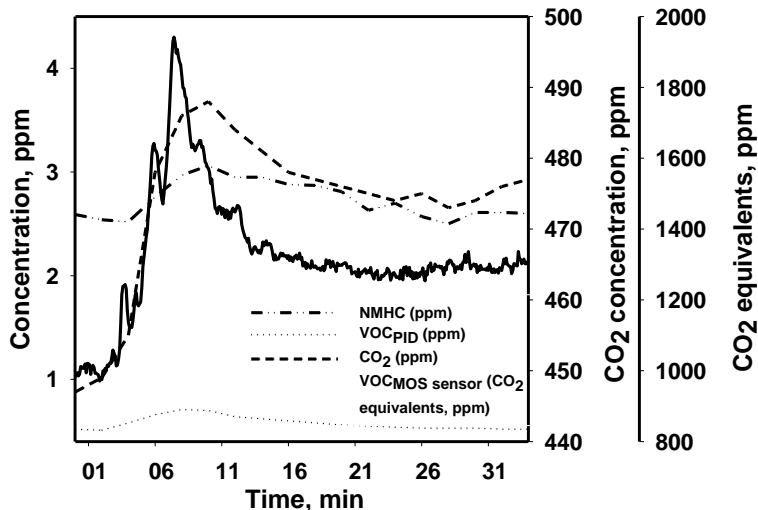

Figure 3.6. Responses and dynamics of gaseous pollutants concentrations in the ventilation exhaust in case of cigarette smouldering.

The correlation analyses between gaseous monitors and simple VOC sensors have revealed that the simple sensors are capable in representing indoor air pollution activities adequately, especially in gaseous phase (VOCs). We found a simple VOC MOS sensor as a very sensitive and responsive measurement technique for representing combined CO_2 and VOC concentrations in indoor air, compared to the PID-based sensor.

The dynamics of indoor pollution sources with the highest registered TVOC concentrations are presented in **Figure 3.7**. The highest TVOCs concentration were registered during the hair spray operation and raised up to 19.16 ppm above the background concentration, followed by the furniture polisher spraying (1.30 ppm), air-freshener spraying (0.81 ppm), oil heating (0.27 ppm), cigarette smouldering (0.19 ppm), and aromatic candle burning (0.16 ppm). The increase of TVOC concentrations were influenced by the following indoor pollution sources as well: incense smouldering (0.12 ppm), water boiling (0.11 ppm), floor mopping with detergent (0.08 ppm), paraffin candle burning (0.08 ppm), and onion frying (0.07 ppm). The TVOCs concentration remained almost unchanged during the following operations: electric stove, radiator and air heater operations, Teflon pan heating, hair dryer, floor vacuuming, and ironing with and without steam operations. TVOC concentrations observed in our study were similar to concentrations obtained in studies from Hong Kong and the United Kingdom (To et al, 2007 and Abdullahi et al., 2013).
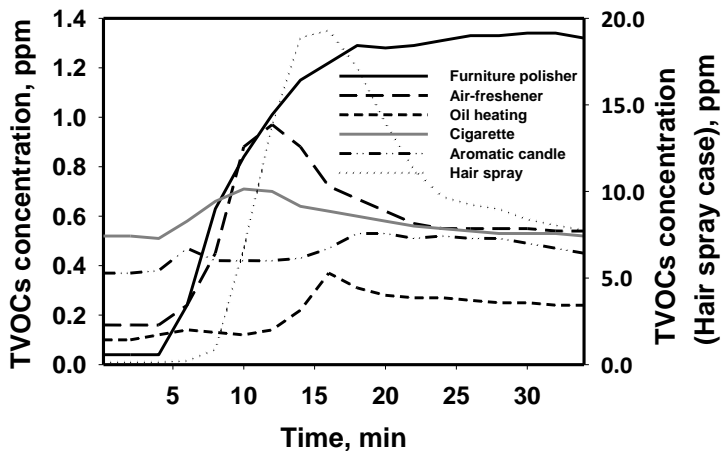

Figure 3.7. Dynamics of indoor pollution sources with highest registered TVOC concentrations.

3.3. IAQ management by combined ventilation and air cleaning

3.3.1. Characterisation of portable air cleaners

The particle size distribution (PNC during peak) as generated by cigarette smoke within a chamber is presented in **Figure 3.8.** Ultrafine particles (0.01–0.42 µm as measured by SMPS) were dominant (dN/dlogDp ranged from ~10^4 to 10^5 cm^{-3}), while larger particles (0.62-2.4 µm as measured by ELPI+) were present at lower

concentration levels ($\sim 10^2$-10^3 p/cm^3). The maximum concentration reached in the chamber was 2.2×10^5 p/cm^3 as measured by SMPS-3910. Generally, the particle size distribution was obtained similar to earlier studies researching cigarette smoke (Daher *et al*., 2010 and Wu *et al*., 2011). Particle size distributions measured by ELPI+ and SMPS-3910 were in close agreement, after converting the aerodynamic diameter to the mobility diameter, even though these instruments measure different physical properties. It must be noticed that SMPS-3910 has been found to yield an error in the range of 0.55-0.97 as compared to a more robust SMPS, attributed to different particle charging technique (Stabile et al., 2014).
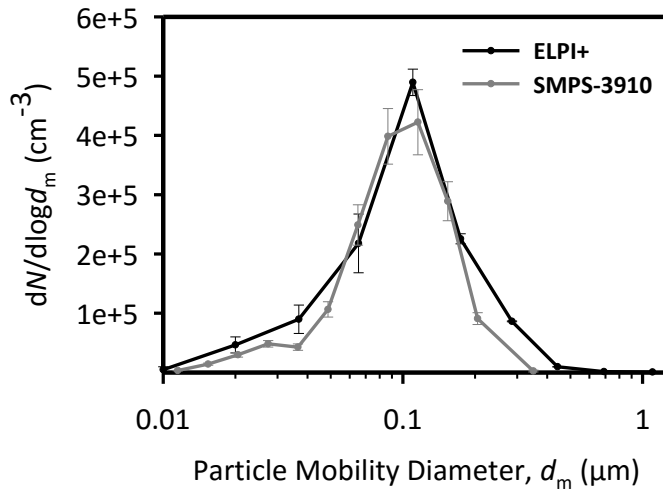
Figure 3.8. The initial particle size distribution of cigarette smoke. Error bars represent the standard deviations of three replicates (dN/dlogd$_m$, particles cm^{-3}).

Table 3.6 lists the main parameters (both declared by the manufacturer and measured ones) of the three air cleaners, including the electrical power draw, air flow rates, CADR$_{PNC}$, CADR$_{VOC}$, E$_{PNC}$, E$_{VOC}$, EEI$_{PNC}$, and EEI$_{VOC}$.

The PAC revealed a relatively large range of measured power draw, i.e. it varied between 6.2 (C1) and 34.7 W (C3) at the low setting and from 15.5 (C1) to 109.1 W (C3) at the high setting. The power draw did not directly correlate with the flowrate among the devices, which varied between 75 (C3) and 126 m^{3}h^{-1} (C2) at the low setting and between 120 (C3) and 276 m^{3}h^{-1} (C1) at the high setting. Different power requirements seemed to be associated with the efficiency of VOC removal, as discussed below. Based on these technical parameters, the tested cleaners were comparable to those used in earlier studies (Mølgaard et al., 2014; Noh and Oh, 2015; Shaughnessy and Sextro, 2006 and Waring et al., 2008).

Table 3.6. Operating modes, operating power (OP), air flow rates, clean air delivery rate (CADR), pollutant removal efficiency (E), and energy efficiency index (EEI) of the tested air cleaners (SD – standard deviation).

Device	Setting	OP^a, W	OP^b, W	Air flow rate^a, m^3h^{-1}	Air flow rate^b ± SD, m^3h^{-1}	$CADR^a$, m^3h^{-1}	$CADR_{PNC}{}^b$ ± SD^a, m^3h^{-1}	$CADR_{VOC}{}^b$ ± SD^a, m^3h^{-1}	$E_{PNC}{}^b$ ± SD^a, %	$E_{VOC}{}^b$ ± SD^a, %	$EEI_{PNC}{}^b$, $m^3h^{-1}W^{-1}$	$EEI_{VOC}{}^b$, $m^3h^{-1}W^{-1}$
C1	LOW	~5.3	6.2	94	97±1.7	Dust 230 Smoke 224 Pollen 245	77±5	2.6±0.4	78.0±1.4	21.4±2.7	14.53	0.49
	MAX	~22	22.8	270	276±3.2		237±11	7.6±1.4	97.2±2.2	33.1±4.9	10.72	0.34
C2	LOW	~10	9.6	130	126±1.2	N/A	106±8	2.2±0.3	81.5±1.6	30.7±2.7	14.58	0.99
	MAX	~16	15.5	210	204±2.4	N/A	153±7	8.2±1.7	90.2±1.8	36.2±5.1	15.62	1.1
C3	LOW	~35	34.7	N/A	75±1.5	~40 m^3/h	37±4	11.2±1.1	53.8±0.9	38.8±4.3	1.05	0.31
	MAX	~100	109	N/A	120±2.8	~82 m^3/h	69±9	19.9±2.8	73.7±1.9	45.7±6.4	0.64	0.19

[a] Manufacturer information
[b] Measured

CADR$_{PNC}$ also ranged widely among the devices and was significantly lower than the air flow rate. The C1 device was the most effective for particle removal (CADR$_{PNC}$ ranging from 77±5 to 237±11 m^{3}h^{-1}). This was supported by high efficiency (E) values (78-97.2%). At the same time, the C3 device was least efficient in particle removal (CADR between 37±4 and 69±9 m^{3}h^{-1}, E between 53.8 and 73.7%). The ratio CADR/Flowrate ranged between 0.49 and 0.85. The CADR and E values for particles were found directly proportional to the air flow rate, with the Spearman rank order coefficient of correlation (r) equalling to 0.83 (p<0.05). At the same time, the CADR$_{PNC}$ was not associated with the power draw, reflecting no association between power draw and flowrate, as mentioned above. The particle removal efficiency was associated with the effectiveness of the installed filter. Only the C1 device has declared the presence of a HEPA class (13) filter, while the other devices did not, and this seemed to have a major effect. On the other hand, the C1 device revealed the discrepancy between flowrate and CADR, considering a very high efficiency of the filter installed. It is not likely that the filter was worn out, since the tested units were new with only several hours of operation. The efficiency of the filter may have been lower than declared, or possibly there might have been a leakage within the unit. At the same time, portable air cleaners were able to reach relatively high particle removal rates in a relatively short period of time (0.5 h from the measured PNC peak value).

The CADR is almost particle size independent in the diameter range from 0.01 to 1.1 µm for cleaners operating at low and max mode (**Figure 3.9**). This is expected because they all draw the air to the filtering system with relatively high filtration efficiency across all particle sizes. Some increase in CADR was noticed in a sub 0.1 µm range for C1 and C3, at low mode as measured with SMPS-3910, possibly reflecting increased collection efficiency in small particles. C1 and C2 operating on the max mode had the highest CADR, with increasing CADR values and particle size. CADR values derived from both ELPI+ and SMPS-3910 were mostly within the margin of error. The values of CADR against particle sizes were of similar magnitude as recently reported by Mølgaard et al. (2014).
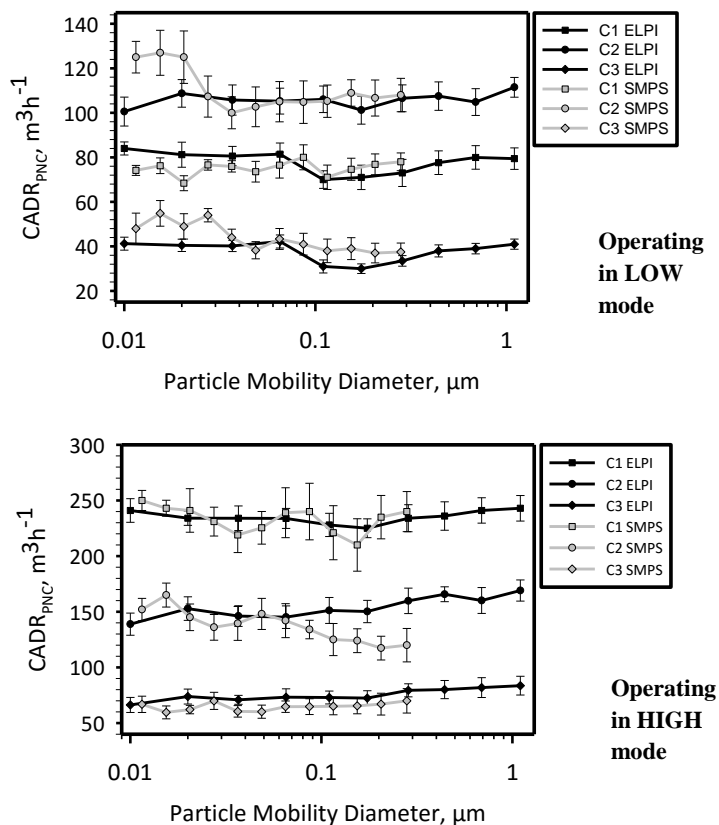
Figure 3.9. CADR$_{PNC}$ as a function of particle diameter in the case of three tested air cleaners (C1, C2, and C3) derived from measurements by ELPI+ and SPMS (Nanoscan). Error bars represent the standard deviation of the CADR for each size bin.

The VOC removal efficiency as reflected by CADR and E was substantially lower, compared to the particle removal efficiency. The cleaner C1 was the least efficient (CADR$_{VOC}$ ranging from 2.6±0.4 to 7.6±1.4 m^{3}h^{-1}, E$_{VOC}$ between 21.4 and 33.1%), while C3 was the most efficient (CADR$_{VOC}$ ranging from 11.2±1.1 to 19.9±2.8 m^{3}h^{-1}, E$_{VOC}$ between 38.8 and 45.7%). The ratio of CADR$_{VOC}$/Flowrate ranged only between 0.02 and 0.16. The CADR$_{VOC}$ and E$_{VOC}$ were not associated with air flow rate but with power draw instead (r=0.94, p<0.05). All three cleaners were based on electrical discharge, implying the decomposition of VOC molecules primarily by ozone. The high decomposition of VOCs may be associated not only with power, but also with the technological arrangements to generate ozone. The configuration of C3 was the most technologically efficient, although associated with a trade-off of higher power requirements. At the same time, the higher power in C3 resulted in efficient decomposition of pollutants, reaching the lowest values of EEI (down to 0.19 m^{3}h^{-1}W^{-1}).

3.3.2. Combined performance of ventilation and air cleaning

This section presents the results from the regression modelling of the effects of simultaneous variation of ventilation and air cleaning intensity to the pollutant removal. The modelling results (response surface plots) of pollutant removal against various ventilation rates and portable air cleaner regimes are presented in **Figure 3.10 (C1), Annex 3 (C2)**, and **Annex 4 (C3).** These plots provide the prediction in CADR, E, and EEI based on the joint variation of ventilation and cleaning intensity.
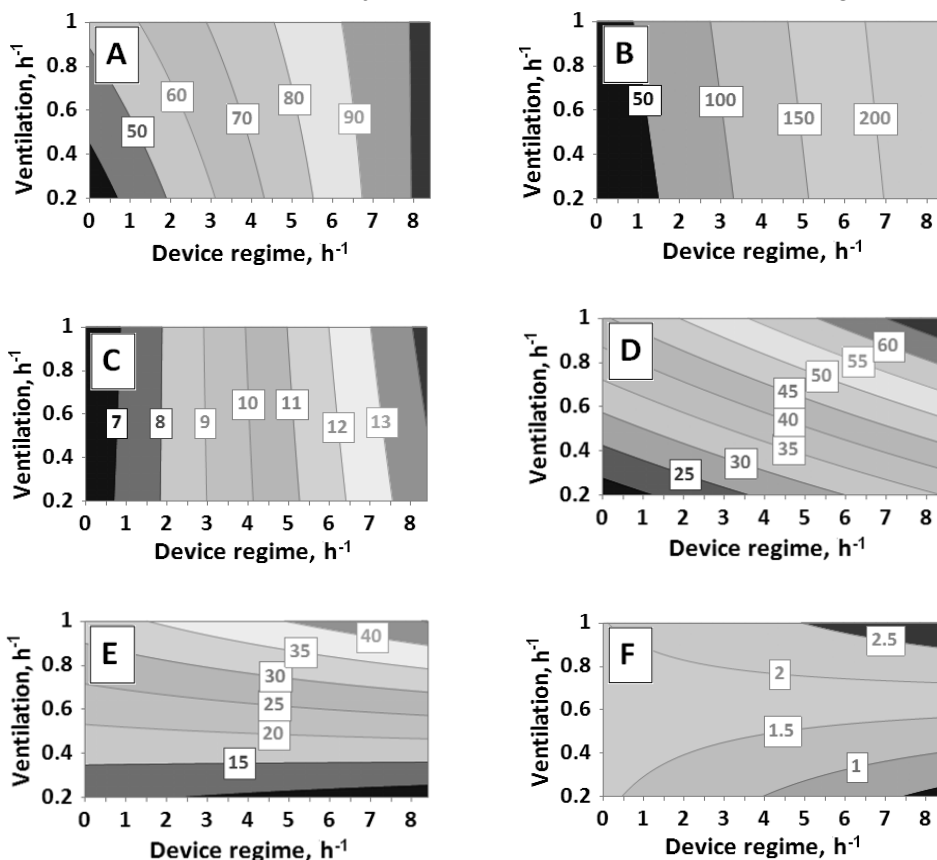
Figure 3.10. Prediction of pollutant removal indicators by covariation of ventilation rates and portable air cleaner (device) regimes in case of cleaner C1: A – E_{PNC} (%); B – $CADR_{PNC}$ (m^3h^{-1}); C – EEI_{PNC} ($m^3h^{-1}W^{-1}$); D – E_{VOC} (%); E – $CADR_{VOC}$ (m^3h^{-1}); F – EEI_{VOC} ($m^3h^{-1}W^{-1}$). The device regime represents multi-pass air changes per hour caused by air cleaner, h^{-1}).

Total decay rate (k_e), including both cleaner and ventilation, decay rate (k_0) of the pollutant concentration, reflecting loss of pollutants due to deposition or adsorption and natural decay rate (k_n), including only ventilation are listed in **Table 3.7**.

Table 3.7. Total (ventilation+cleaning) decay rate (k_e), natural (only ventilation) decay rate (k_n), and loss (deposition+adsorption) decay rate (k_c), as calculated from the measurements in each scenario.

	Device	Device operating mode	PNC Ventilation 0.2, h^{-1}	Ventilation 0.6, h^{-1}	Ventilation 1.0, h^{-1}	VOC Ventilation 0.2, h^{-1}	Ventilation 0.6, h^{-1}	Ventilation 1.0, h^{-1}
k_e	C1	LOW	2.9±0.1	3.2±0.1	3.4±0.1	0.9±0.1	1.3±0.1	1.7±0.1
		MAX	7.3±0.3	7.6±0.4	7.8±0.4	1.0±0.1	1.4±0.1	1.8±0.1
	C2	LOW	3.7±0.1	4.0±0.2	4.2±0.2	0.8±0.1	1.2±0.1	1.6±0.1
		MAX	5.0±0.2	5.3±0.2	5.5±2.4	1.0±0.1	1.4±0.1	1.8±0.1
	C3	LOW	1.8±0.1	2.1±0.1	2.3±0.1	1.1±0.1	1.5±0.1	1.9±0.1
		MAX	2.7±1.1	3±0.1	3.2±0.2	1.3±0.1	1.7±0.1	2.1±0.1
k_n			0.5±0.1	0.8±0.1	1.1±0.1	0.5±0.1	0.9±0.1	1.3±0.2
k_c			0.3±0.1	0.2±0.1	0.1±0.1	0.3±0.1	0.3±0.1	0.3±0.1

The removal of particles from the indoor air as represented by E_{PNC} was affected by both ventilation and air cleaning. Such phenomenon was valid for all tested devices (**Figure 3.10A**, **Annex 3A** and **Annex 4A**). The removal of particles seemed to be more affected by the increase in ventilation. For example, the increase in E_{PNC} from 40 to 50% would be achieved by changing ventilation rate from 0.4 to 0.9 h^{-1} with no air cleaner in operation. The same efficiency would be reached by increasing cleaner flow rate from 0.7 to 1.9 h^{-1} with minimum ventilation. The E_{PNC} was at ~55% in case of maximum mechanical air ventilation rate (1 h^{-1}) and with no PAC operating. In the opposite setting, i.e. at the lowest setting of ventilation (0.2 h^{-1}), the E_{PNC} of 55% may be achieved at the device regime of ~2.5 h^{-1}. While the ventilation seems to be more efficient, the ranges of its operation in residential buildings are rather constrained and go up to 2 h^{-1} (except of kitchen hoods). Thus, the high E_{PNC} values may not be achieved using ventilation alone, or would take prolonged periods of time. The cleaners become more efficient at the higher flowrates, producing flows equivalent to – 4 h^{-1}. Such flowrates allow efficiencies of 75-90% to be achieved during a period of 30 min even at the minimum ventilation. Moreover, the model suggests that at the highest air cleaner flow rates the effect of ventilation is minimised, as represented by isolines almost perpendicular to abscissa axes (**Figure 3.10A**, **Annex 3A**, and **Annex 4A**).

The $CADR_{PNC}$ appeared to be even less affected by the ventilation compared to the air cleaning (**Figure 3.10B**, **Annex 3B**, and **Annex 4B**). The effect of ventilation was only noticeable in the case of device C3 having the lowest efficiency in particle filtration and the lowest CADR. Similar findings extended to the energy efficiency index for PNC (EEI_{PNC}). The EEI_{PNC} did not depend on the changes in ventilation rate (except C3) but it increased with increasing device flowrate. This

was due to the fact that increasing flowrate did not cause a dramatic increase in the energy consumption of the tested devices C1 and C2. C3 showed the lowest energy efficiency which was comparable to that of ventilation. These findings indicate that in areas where cleaners will primarily be used for the removal of particles, the EEI should be observed while selecting a cleaner; these metrics must be provided by the manufacturers. In cases of particle removal in residential areas the strategy of the lowest ventilation coupled with the highest air cleaner operation rate may yield the best results in terms of particle removal efficiency during the shortest amount of time and the lowest energy consumption.

It must be noted that the above results were obtained using a certain configuration of filtering efficiency in single-pass (ventilation) and multi-pass (cleaners) processes. Our ventilation supply was equipped with a very efficient HEPA filter, which is not commonly used in dwellings. The penetration of outdoor particles and particle size distribution in the single-pass process greatly depend on a filter class installed (Azimi et al., 2014), while the multi-pass process can compensate for the lower efficiency by providing multiple circulation of air through the filter. Thus, having a filter of a lower efficiency at the air handling unit (such as MERV13) may provide a higher impact of air cleaners towards the removal of indoor aerosol. On the other hand, if the efficiency of the air cleaner filter is boosted by charging (electret based filtration), it may decrease through the operation span and also decrease the cleaner performance.

The removal of VOCs in a chamber appeared to be affected more by the ventilation than the air cleaning, as indicated by horizontal direction of regression lines in surface plots. This corresponds to the PAC test results, where all tested cleaners scored relatively low in VOCs removal efficiency. The ventilation rate of 1 h^{-1} alone was able to achieve the E_{VOC} at ~40-45%, while similar efficiency E_{VOC} is achieved at 3.5 h^{-1} of cleaner intensity (at a minimum ventilation, C3, **Annex 4D**) and ~8-9 h^{-1} (C1 and C2, **Figure 3.10D** and **Annex 3D**, respectively). The opposite strategy to the particle removal would include using no air cleaning and maximum ventilation. This would allow pollutant removal efficiency of 40-45% to be achieved within 30 min. Adding air cleaning at the lowest setting would increase removal efficiency by ~5%, while additional air cleaning at the maximum flowrate would result in a removal efficiency of ~60% in 30 min.

The $CADR_{VOC}$ curve between the ventilation and air cleaner device regimes is nearly horizontal, showing little impact of air cleaning to the removal of VOCs as compared to the ventilation (**Figure 3.10E**, **Annex 3E**, and **Annex 4E**). Meanwhile, the removal of VOCs by joint ventilation and air cleaning seems to be the most energy-efficient strategy at the maximum mechanical ventilation rate (1 h^{-1}) and maximum device regime, except of the device C3, which was not energy efficient at high flowrates. Similarly, to our findings, it has been indicated that efficiencies of gaseous pollutant cleaners vary greatly from product to product and from VOC to VOC (Chen et al., 2005 and Kim et al., 2012). Our surrogate pollution source also contained specific VOCs which may be decomposed differently in other types of air cleaners.

The above presented results indicate that air cleaners, although not very efficient, may add additional value for the removal of VOCs in indoor air in order to have a faster and energy efficient reduction of pollutant concentrations. At the same time there are doubts associated with the ozone-based decomposition of VOCs, which may release partial decomposition products to indoor air that may be of even higher toxicity than the original VOCs (Waring et al., 2008). Moreover, the release of ozone (if any) may affect human health directly or via the formation of nanoparticles during reactions with VOCs occurring in air (Chen et al., 2005). In this study, we did not register ozone levels above the detection limit of the instrument, nor did we detect a smell of ozone. On the other hand, the decreasing quality of outdoor air in specific pollution episodes may not provide suitable air to dilute the indoor VOCs. Thus only well designed and balanced ventilation providing a good quality air may be a more feasible strategy for the removal of VOCs from the air (together with other gaseous pollutants, such as CO_2). More advanced IAQ management can be based on the real-time monitoring of indoor and outdoor air and the adjustment of ventilation/air cleaning strategies based on the real time air quality data.

3.4. A grey model to IAQ management in rooms based on real–time sensing of particles and VOC

3.4.1. Temporal variation of pollutant concentration in room and source strength

The increase/decrease phases of pollutant concentration in a room during various pollution episodes were approximated by sigmoidal and exponential decay functions, respectively (**Methods section**). The tested indoor air pollution sources have caused distinctly different variations in pollutant concentrations in time. Some pollution sources, such as cooking were distinct for their fine particle emission; others, such as cooking oil heating on an electric stove, Teflon pan heating on an electric stove, and incense and cigarette smouldering showed coarse particle emission, as determined by the nephelometer. Most of the pollutants were associated with VOCs emission, however, the intensity in temporal variation of pollutant concentration was significantly different. The pollution sources were arbitrarily distinguished to fast and slow, depending on their aerosol emission (Ciuzas et al., 2015); the same can be applied to the VOC concentrations in rooms. This study aimed at the utilisation of this data for the further modelling of the ventilation-based IAQ management.

The above described functions were further approximated using Equation (14). These approximations served as input data for Equation (15) for estimating the variation in source strength based on fine, coarse particles and VOC concentrations. An example of such source variation in case of coarse particles emitted from Onion frying in oil in a Teflon pan is presented in **Figure 3.11**.
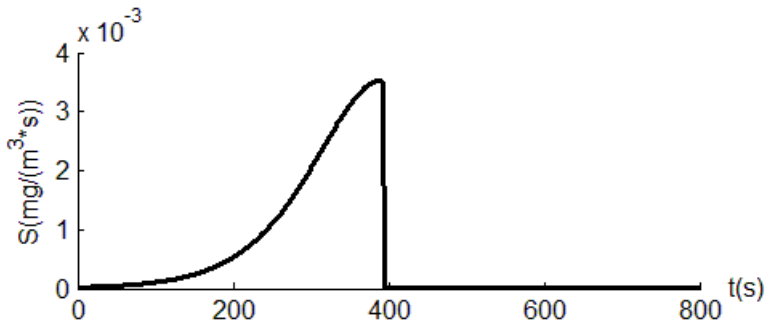
Figure 3.11. The modelled variation of emission of coarse particles during onion frying operation.

This figure indicates that pollution sources are not constant in time. The release of fine particles increases with time (start-up time t = 0 s), and decreases gradually after the process has been terminated. Similar results have been obtained in case of other sources as well, namely, the pattern of temporal variation was varying among sources, but the sources were not constant.

3.4.2. Construction of the model for IAQ management

The model for the IAQ management based on the real time variation of fine and coarse PM as well as VOC concentration is comprised of three blocks, each representing a specific pollutant. These three blocks provide an output to the ventilation rate controller block, where calculated levels of pollution are compared with set threshold values (**Figure 3.12**). The ventilation rate λ is being changed according to the result of the comparison. Although this model focuses on the specific pollutants originating from indoor activities, it may be amended by the measurements of other common IAQ parameters, namely CO_2 and relative humidity.
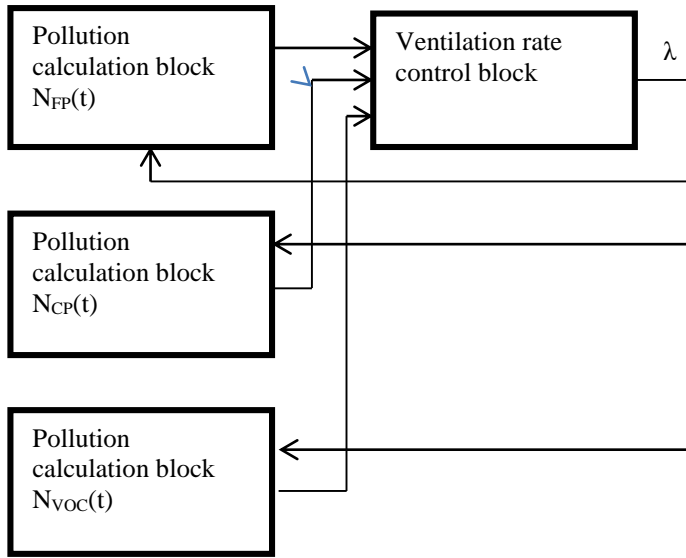
Figure 3.12. The structure of the IAQ management model (FP – fine particles, CP – coarse particles, VOC – volatile organic compounds, N(t) – concentration in time).

Multiple algorithms have been applied for the management of HVAC systems within homes, including classical PID regulators, Fuzzy logic regulators, neural-network based regulators and various modifications of the above (Afram and Janabi-Shafiri, 2014b). In our case, these algorithms have certain limitations. For example, Fuzzy logic requires very specific knowledge on the structure of the building and its conditions. Neural networks require an extensive data set for training of the model. PID regulators are some of the most practical when applied to the ventilation systems, based on the energy consumption as an optimising criterion. The following equation is being applied (Chao and Hu, 2004).

$$\lambda[i] = k_p \Delta N[i] + k_d \left(\Delta N[i] - \Delta N[i-1]\right) + k_i \sum_{j=1}^{i} \Delta N[j],$$

$$\Delta N[i] = N[i] - N_{sp}$$

(20)

Where: $N[i]$ – is air pollutant concentration in time i; k_p, k_i, k_d – proportional, integral and derivative component; N_{sp} – threshold value of pollutant.

Our aim was to minimise the time during which the pollutant concentration falls below the threshold value. Thus, on the opposite of the PID regulation, the supply air rate should always be increased if the pollutant concentration grows, even if it is below the threshold value. Furthermore, the ventilation rate should be reduced once the pollutant concentration decreases and reaches the threshold value. The integral component must be omitted, since it slows down the reaction of the system to the changes in air pollutant concentration.

The ventilation rate is being controlled according to the following expression:

$$\lambda_i = \lambda_{i-1} +$$
$$\Delta\lambda\frac{1+\text{sgn}(\Delta N_m - \Delta N)}{2}\text{sgn}(\Delta N_m - \Delta N)\frac{1+\text{sgn}(\lambda_{max} - \Delta\lambda - \lambda_{i-1})}{2}\text{sgn}(\lambda_{max} - \Delta\lambda - \lambda_{i-1}) +$$
$$\Delta\lambda(1-\text{sgn}\,\beta)\frac{1+\text{sgn}(\lambda_{max} - \Delta\lambda - \lambda_{i-1})}{2}\text{sgn}(\lambda_{max} - \Delta\lambda - \lambda_{i-1}) -$$
$$\Delta\lambda\frac{1-\text{sgn}(\Delta N_m + \Delta N)}{2}\text{sgn}(\Delta N_m + \Delta N)\beta \qquad (21)$$

Where: $\Delta\lambda$ is the step of the ventilation change rate (s^{-1}), λ_{max} is the maximum possible value of λ (s^{-1}), $\Delta N_{FP} = N_{FP}(t_{i-1}) - N_{FP}(t_{i-2})$, $\Delta N_{CP} = N_{CP}(t_{i-1}) - N_{CP}(t_{i-2})$, $\Delta N_{VOCP} = N_{VOC}(t_{i-1}) - N_{VOC}(t_{i-2})$ is the change of the fine particle, coarse particle and VOC concentration, respectively (increase or decrease), ΔN is the maximum change of the fine particle, coarse particle and VOC concentration (#/cm^3 for fine PM, mg/m^3 for coarse PM, ppm for VOC), ΔN_m is the threshold value (ventilation rate is increased when ΔN exceeds this threshold value) (#/cm^3 for fine PM, mg/m^3 for coarse PM, ppm for VOC), β is the condition that none of the pollutant concentrations exceed threshold values.

$$\beta = \frac{1+\text{sgn}(N_{FPn} - N_{FP})}{2}\text{sgn}(N_{FPn} - N_{FP}) \times \frac{1+\text{sgn}(N_{CPn} - N_{CP})}{2}\text{sgn}(N_{CPn} - N_{CP}) \times$$
$$\frac{1+\text{sgn}(N_{VOCn} - N_{VOC})}{2}\text{sgn}(N_{VOCn} - N_{VOC}) \qquad (22)$$

Where: N_{FP}, N_{CP} and N_{VOC} are the concentration levels of fine particles, coarse particles and volatile organic compounds (#/cm^3 for fine PM, mg/m^3 for coarse PM, ppm for VOC), N_{FPn}, N_{CPn} and N_{VOCn} are the appropriate threshold values (#/cm^3 for fine PM, mg/m^3 for coarse PM, ppm for VOC).

Thus, the workflow of the entire modelling process is summarised as follows:

1. The variation of pollutant concentration in a chamber was measured at constant air ventilation rate;

2. The variation of pollutant concentration was approximated by a function f(t) (14);

3. The variation of pollution source intensity *S(t)* was calculated (**ANNEX 5b**);

4. The air change rate was varied using (21) in order to minimise the pollutant concentration, which was modelled using differential equation (19) and using *S(t)* is used as the input in the model (**ANNEX 5c** and **Figure 3.13.**).

The suggested management algorithm is different from the standard PID algorithm due to introduced non-linearity; the ventilation rate is not decreased but rather increased as long as one component exceeds the threshold values. Moreover, in this algorithm the air flow is being changed by a constant value of $\Delta\lambda$, since the rate of pollutant concentration variation is low and the ventilation rate adjustments are discrete, thus the system has time to adjust itself. The algorithm was also tested at the condition of $\Delta\lambda = k\Delta N_m$, however it did not have a substantial effect.

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3.4.3. Results of the modelling

The researched pollution sources were grouped into two groups; food preparation (including incense smouldering) sources and "non-food" sources. This has been done in association with the limits of ACR that was available to vary. In kitchens air exhaust hoods can create much higher ACR. Thus, the following maximum ventilation rates were used: first group $\lambda_{1max} = 20$ h^{-1} and $\lambda_{2max} = 4$ h^{-1}. The usual ACR in rooms varies around 0.5 h^{-1}. The following threshold values were chosen: N_{FP} – 10000 #/cm^3, N_{CP} – 0.05 mg/m^3, N_{VOC} – 1000 ppm CO_2 equiv.

The modelling results are presented as concentration variations in time (**Figure 3.14.**). Since all of the researched pollution sources have their specific emission pattern of VOCs and particles, the model addresses the pollutants which have a pronounced variation. Incense smouldering mostly releases fine particles (**Figure 3.14A**), thus ventilation rate is adjusted according to this pollutant (**Figure 3.13**). In case of a ventilation rate of 4 h^{-1}, the threshold value is reached after 25 minutes from the start of the pollution episode. If the ventilation rate was kept as at 1 h^{-1}, the threshold value would be reached in 105 minutes. Although ventilation standards do not regulate the time to achieve good IAQ from the start of the pollution episode, such real time modelling and adjustment of ventilation rate would optimise the performance of air handling unit.

Some pollution sources are difficult to efficiently manage even by maximum ventilation. For example, coarse particle concentration in the case of aromatic candle is reduced only after 64 min at the ventilation rate of 4 h^{-1} (**Figure 3.14B**), while fine particle concentration stays elevated in the case of hair spray for 120 min and longer (**Figure 3.14C**) or the hair dryer (**Figure 3.14D**). Fine particles are a specific pollutant which can form as a result of the reactions of VOCs and ozone. The air handling units in homes are usually limited to 1-2 air changes per hour since it is not economically feasible to install larger units. In such cases, some pollution episodes may affect IAQ for prolonged periods of time. It may be suggested to shift such activities to zones having higher ventilation rate, i.e. kitchens. Some modelled sources were low emitting (such as electric radiator), which did not contribute to pollution episodes approaching threshold values (**Figure 3.14E**).
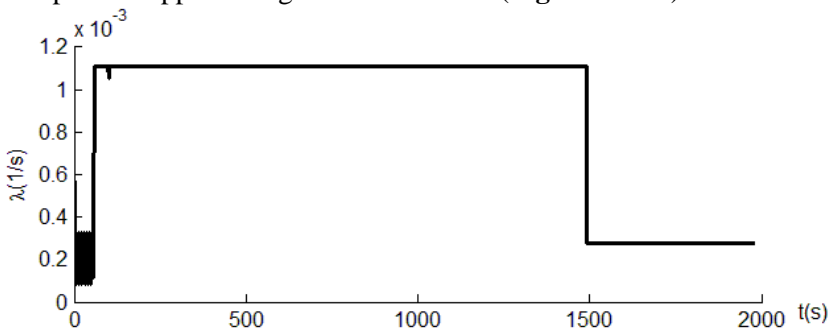

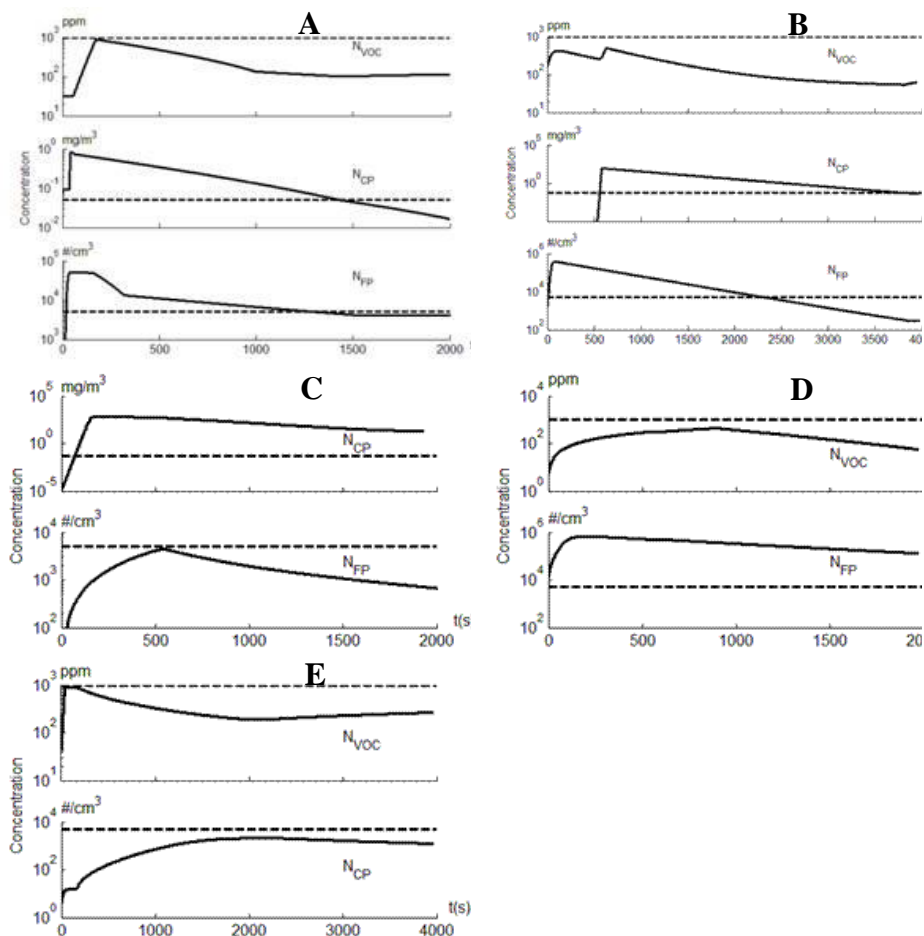**Figure 3.14.** Variation of ventilation rate in case of incense smouldering.

Figure 3.15. The variation of VOCs, fine particle and coarse particle concentrations in time after the application of air quality management model at a set ventilation rate and threshold values of pollutant concentration (dashed lines). A) incense smouldering; B) aromatic candle; C) hair spray; D) hair dryer; and E) electric radiator.

Based on the above presented modelling results, the "non-food" pollution sources were divided to three groups (**Table 3.8**):

- High-intensity sources, causing (relatively) long term pollution exceeding threshold values;
- Medium-intensity sources, causing short term exceedances of threshold values;
- Low-intensity sources that do not cause a worsened IAQ.

The table illustrates the emission patterns from different sources and the necessity for measuring selected analytes in order to effectively mitigate their impact to the IAQ.

Table 3.8. The classification of pollution sources based on their intensity and emitted pollutants.

Type of pollution source	Pollution source	Pollution exceeding rates
High-intensity sources	Hair spray	N_{CP}
	Hair dryer	N_{FP}
Medium-intensity sources	Incense smouldering	N_{FP}, N_{CP}
	Air-freshener (aerosol)	N_{FP}, N_{VOC}
	Candle burning	N_{FP}, N_{CP}
	Furniture polisher	N_{CP}
	Floor wet mopping (detergent)	N_{VOC}
	Ironing with steam	N_{CP}
Low-intensity sources	Floor vacuuming	-
	Ironing without steam	-
	Electric radiator	-
	Electric air heater	-

Food preparation sources as well as cigarette burning are very intense sources, thus, a ventilation rate of $\lambda_{max} = 4$ h^{-1} was found to be too low and the ventilation rate of 20 h^{-1} has been employed. In fact, kitchen exhaust hoods have a high potential to serve as ventilation devices in premises where other ventilation systems are unavailable.

At $\lambda_{max} = 20$ h^{-1}, water boiling, oil heating and onion frying can be attributed to medium intensity sources. At the same time, cigarette burning was the intense source, causing the exceedance of threshold values after 3 hours.

An additional modelling case was selected to test the importance of the measurement of fine particles; fine vs. coarse (**ANNEX 6**, onion frying). In case the ventilation rate is managed only by the measurement of coarse particles (the concentration of fine particles is not used to manage the ventilation rate), the concentration of coarse particles is reduced in a 10 min period to acceptable levels. However, the concentration of fine particles stays elevated. This illustrates that ventilation systems operating on the data produced by nephelometer may not be effective to assure healthy indoor air. Efforts in introducing inexpensive nanoparticle monitors to ventilation systems should be taken by the manufacturers of particle measurement systems.

The above presented modelling and control approach may have several limitations. Firstly, the grey box model has been developed and implemented using data obtained in controlled conditions, i.e., simulating pollution episodes in an environmental chamber. Such set up was developed in order to simulate real-life situations, but the amount of degrees of freedom in real living environments is much larger, including poorly controlled ventilation rates, different emission characteristics of particular sources, the dispersion of pollutants in particular

settings, etc. Moreover, there may be multiple pollution sources operating simultaneously. The proposed algorithm aims at taking advantage of a general control concept that the supply air rate should be always increased if the pollutant concentration grows, even if it is below the threshold value. In this way, uncertainties associated with varying pollutant emission patterns or multiple sources may be tackled, while only the time to reduce the air quality below threshold value may be different. In this perspective, the suggested algorithm is different from the industry-standard proportional-integral-differential (PID) management algorithm, which is usually applied for the control of IAQ based on temperature readings. The developed grey box model may be attributed to a hybrid model group, as recently summarised by Okochi and Yao (2016), presumably providing a more robust algorithm, although an additional effort for the development has been spent including physical laboratory simulations. At the same time, this model does not require adaptive learning techniques to be applied, which may be of an advantage, since current practices are not always able to address non-linear processes that occur in buildings (Huang et al. 2010).

Another issue that may be encountered during the implementation of this algorithm is the uncertainties associated with the robustness of sensors used for measurements of IAQ. The CO_2 sensors have been shown to be prone to uncertainties, plus they require constant checks and calibration (Okochi and Yao, 2016). Another major problem is sensor noise, which may provide false signals for the model to judge the changes in pollutant concentration that need to be addressed. Specific signal processing filters may be considered and implemented to eliminate the effects of noise in order to have a reliable operation.

4. DISCUSSION AND RECOMMENDATIONS ON THE APPLICATION OF THE FINDINGS OF THE RESEARCH

4.1. Indoor air quality and source control

4.1.1. Findings on the IAQ in low energy buildings in Lithuania

The research shows that IAQ in the investigated 11 low energy buildings was generally good, except of a few analytes, which provide room for improvement. The ACR in most buildings was rather low (0.08-0.69 h^{-1}). This may be a key feature of Lithuanian low energy buildings, where ventilation systems are not properly installed and balanced, or residents lack the knowledge on the management of ventilation to meet the requirements for IAQ. Such phenomenon indicated an opportunity for the accumulation of pollutants released indoors. While CO_2 and many tested VOC and SVOC concentrations were at reasonable levels, the concentration of formaldehyde was elevated above the national limit values (3.3-52.3 µg/m^3). Apparently, the emission rate of formaldehyde from multiple sources was higher than the forced ventilation systems operating at low intensity were able to extract.

In several buildings, extremely high concentrations of VOCs were observed where the interior installation activities were done just prior to the measurement

campaign. The decrease of BTEX concentrations was rapid and fell below limit values in one month. This relatively fast rate may be attributed to the steady ventilation of 0.5 h^{-1} which facilitated the removal of VOCs.

The presented research results indicate that adequate mechanical ventilation must be assured in low energy buildings. While low occupancy of single family buildings may be beneficial to keep most of the pollutants within the recommended values, stronger emitting sources may be difficult to manage, especially immediately after installation. Checking of IAQ is recommended before occupancy to avoid moving into buildings before the complete installation of the interior. Moreover, the concept of low energy buildings should be coupled with a "healthy and sustainable building" concept, aiming to avoid usage of highly emitting building and furniture materials.

4.1.2. Findings of characterisation of active indoor pollution sources and their impact on IAQ

The tested groups of indoor air pollution sources by uniform aerosol measurement methods have revealed a broad range of particle concentrations. The hair dryer operating, Teflon pan heating, and cooking oil heating were observed to be the strongest of the $PM_{0.01-0.3}$ sources (with maximum concentrations of 4.2×10^6 p/m^3, 2.6×10^6 p/m^3, and 1.6×10^6 p/m^3, respectively), followed by the candle burning, cigarette and incense smouldering, frying, cooking, and aerosol spraying. The lowest concentrations of particles were registered during the operation of electric radiators, electric air heater, electric stove, ironing and floor vacuuming, resulting in concentrations of 10^3 p/m^3 order of magnitude.

The operation of most sources revealed significant temporal variation. The pollution sources were arbitrarily classified to a rapid and slow increase on PNC based on their temporal profiles. Some sources such as candle burning and cigarette smouldering displayed a rapid increase and a relatively fast decrease of concentration. Others showed a rapid increase (such as aerosol sprays) and low decrease, due to secondary aerosol formation. Such classifications of pollution sources provide data for the air quality management algorithms on setting values for air replacement or cleaning intensity.

Two locations within a room were tested for IAQ monitoring based on aerosol concentration, including in the duct of the exhaust ventilation and the centre of the ceiling. Both locations proved to adequately represent the concentration levels and dynamics of pollution episodes.

The variation of PM concentrations in the size range from 0.01 to 0.3 mm reflected the highest complexity with respect to the temporal variation, mainly due to complex mechanisms of particle formation and transformations. Furthermore, the rapid variation of concentrations due to turbulent dispersion regimes may cause difficulties to IAQ management algorithms aiming at controlling IAQ based on the growth and decay rates of concentrations.

The highest TVOCs concentrations were registered during the hair spray operation and raised up to 19.16 ppm.

Thus, the combination of air cleaning and ventilation may be an efficient solution for a more efficient IAQ management provided that the co-operation of these devices is well considered and managed according to measurements of specific pollutants.

4.1.3. Findings of IAQ management by combined ventilation and air cleaning

The performance of three portable air cleaners was evaluated in the test chamber by examining tobacco smoke concentrations. The tested cleaners are effective in the removing of indoor tobacco smoke particles, reaching 97% particle number removal efficiency after 30 min. The VOCs removal efficiency was lower and varied from 21.4 to 45.7%. $CADR_{PNC}$ varied widely among tested cleaners and their operation intensities, and ranged from 37 (±4) to 237 (±11) m^3h^{-1}, while $CADR_{VOC}$ ranged from 2.2 (±0.3) to 19.9 (±2.8) m^3h^{-1}.

Generally, PAC proved to be more efficient devices than the ventilation to remove aerosol particles. The maximum air cleaning and minimum ventilation allowed efficiencies of 75-90% to be reached during 30 min as well as the highest energy efficiency values. With VOCs, ventilation with no air cleaning removed 40-45% of VOCs, and combined with air cleaning the removal efficiency topped at ~60%, however, not taking into account the release of the decomposition sub-products. These results provided data for the consideration for the real-time IAQ management based on measurements of aerosol particles and VOCs indoors.

4.1.4. Findings of grey model approach to IAQ management in rooms based on real-time sensing of particles and VOC

The indoor pollution sources have very distinct patterns with respect to VOCs and PM emission, thus all of the suggested indictors must be measured and modelled. The created grey box model for the management of IAQ based on real-time measurements of VOCs and fine and coarse particles utilises a novel approach to the IAQ management algorithm and can be effectively applied to the control of air handling units in homes. The model can be amended by the measurement data of other pollutants in a similar manner as demonstrated, as well as manage a combination of ventilation and air cleaning systems. Such applications will assure an optimal utilisation of the air handling unit in order to achieve the acceptable IAQ in the lowest time span and optimal energy use. In case of food preparations and other very intense sources, only the high ventilation rate (such as available in a kitchen exhaust) may assure an adequate time span to reach the acceptable IAQ.

4.2. IAQ management system

4.2.1. Air pollutant measurement technology

The efficient management of indoor environmental quality should not only rely on the control only of temperature, relative humidity and concentration the CO_2, because these parameters do not fully represent indoor air pollution. The airborne

pollutants indoors are found in both gaseous and particulate phases. It is recommended to utilise real-time measurements of PM (both nano and micro ranges), and VOCs for the control of IAQ. There are many different PM measurement techniques on the market, but only a few of them have the potential to be integrated into IAQ control systems. The main criteria include cost of the device, maintenance requirements and noise emissions during the operation.

The review of the measurement techniques revealed that nephelometery may be the most suitable technique to measure indoor micro particle concentrations. The lower measurement range starts at 0.3 µm, thus, not being able to register nanoparticles, and this is the main disadvantage of this technique. At the same time, no such low cost solutions have yet been developed for the nanoparticle measurements. It is believed that low-cost and simple to operate nanoparticle counters will be widely available soon due to the intensive research in this area.

The indoor concentrations of VOC can be measured using the real time meters based on the photo ionisation detector (PID) as a VOC sensor. In these sensors, the VOC concentration is expressed as the total organic carbon concentration that represents the equivalent of a certain mixture or a single VOC that it was calibrated against. The PID is a relatively costly technique and requires constant maintenance, thus making it unattractive for installations in homes.

Semiconductor or Metal Oxide Sensors (MOS) are one of the oldest and least expensive measurement technologies used in portable instruments. The semiconductor technology based MOS sensors are spreading in the marked as an alternative for VOC concentration measurement. They are compact and low cost sensors. These sensors are suitable for the detection of various non-methane hydrocarbons, including aliphatic and aromatic hydrocarbons, alcohols, ketones, organic acids, amines, etc. These sensors are cheap and provide good time resolution and sensitivity. They may also be calibrated to convert VOC measurement results into an equivalent CO_2 concentration, thus achieving compatibility between these measurement results and the CO_2 concentration standards. At the same time, these sensors still suffer from baseline drift and too short a service life to rely on them for a long-term operation. Tests performed on some Figaro MOS sensors found that they had more drift during exposure to xylene than the polymer-absorption sensors. While MOS sensors can detect a very wide range of contaminants, they have number of shortcomings that limit their effective:

- They have limited sensitivity, with detection limits usually in the 10's of ppm;
- They produce a non-linear output so they are not particularly accurate. MOS sensors are at best a gross indicator for toxic gases and vapours;
- They are slow to react (relative to a PID);
- They respond positively to moisture and temperature;
- They can be poisoned and dirtied and are not easily cleaned;
- MOS sensors are the first true "broad-band" sensors so they respond to a wide variety of compounds.

4.2.2. Positioning of PM and VOC sensors in buildings

An important consideration is the positioning of the monitoring devices within a room. Depending on the ventilation strategies, the pollutants may be dispersed differently. It is almost impossible to assume a well-mixed state within a room, thus there may be a certain uncertainty introduced while selecting a place for the monitoring device.

Based on the study results two strategies are recommended, based on the type of ventilation:

1) In the case of forced ventilation, air quality monitors may be installed in the exhaust ventilation duct. This has several advantages. Firstly, the pollutants from a room are concentrated and directed into a single air stream, where an integrated sample of all the air in the room can be taken. Secondly, the mechanical ventilation unit already provides the airflow required for passing a sample of air through the sensor, thus the pump less design may be effectively utilised. We believe that in increasing energy requirements for buildings, the mechanical ventilation systems will prevail in most of the buildings, thus monitoring strategies can be focused towards the monitoring of ventilation air.

2) In situations without mechanical ventilation, a "smoke detector" approach may be employed, where monitoring sensors are positioned on the ceiling in the centre of the room. This poses some challenges in already inhabited spaces, since there may be no possibility to direct power cables to such location.

4.2.3. Displacement vs cleaning of pollutants

Low-energy houses rely on forced mechanical ventilation. Mechanical ventilation provides ventilation conditions, when used indoor air is removed and at the same time replaced by outdoor air. In ventilation systems air may be additionally filtered, heated or cooled, moistened or dried. This is usually installed in air handling units.

In areas where the outdoor air is severely polluted, the direct supply to indoor air may worsen the indoor air quality. In such case, pre-treatment of outdoor air should be conducted, which includes not only filtration, but also removal of gaseous pollutants. Such capabilities may be provided by an in-ducts air cleaning device, which not only effectively filters the outdoor air, but is clean of VOCs and/or other gaseous pollutants. Various air treatment technologies have emerged in the market, including plasma treatment (advanced oxidation), catalysis, UV radiation, filtration, sorption, etc.

For the treatment of pollutants originating within a building, ventilation may not be sufficient due to restricted air change capacity, usually limited to 1 ACH. With strong indoor sources of pollution and insufficient ACR the emitted pollutants result in a worsened IAQ. In order to minimise the effects of indoor air pollution sources impact on IAQ and to save the energy required for ventilation, portable air cleaners may be utilised. Such air treatment devices are also based on several parallel technologies (filtration, plasma, catalysis, etc.), and are capable of the efficient removal of PM and to some extent VOCs within a short period of time. Our

research found that these two technologies can be effectively integrated to maximise their efficiency.

4.2.4. IAQ control algorithms

The existing heating, ventilation and air-conditioning algorithms are based on real-time sensing of control parameters. Various modelling approaches have been utilised to predict and manage such systems, including white box, black box, grey box, sub-zonal and multi-zone, Computational Fluid Dynamics, and Windows-based IAQ simulations. White box models are entirely based purely on the deterministic equations representing physical processes. If these are not entirely known (which is usually the case with complex systems such as indoor air), the relationships between input and output signals is approximated using experimental data (measured input and output signal values). In grey box models, analytical solutions to the equations of the processes are known while experimental data is used only for the determination of some parameters for analytical solutions.

The designed IAQ management model, based on fine and coarse particle concentrations, and the VOC concentration measurement in real time, is composed of three blocks, each representing a different pollutant. These three blocks send a signal to the air change control unit to set the pollution level and it is compared with predefined threshold values. The ACR is changed according to the received data comparison results. Although this model focuses on the specific pollutants originating from indoor activities, it may be amended by the measurement of other common IAQ parameters, namely CO_2 and relative humidity.

4.2.5. Integration of proposed technologies to a building IAQ control system

The recommended scheme of the IAQ control is presented in **Figure 4.1.** This scheme consists of the following segments: sensors of pollutants; air handling unit; in-duct air cleaning device; portable air cleaning device; automatic air flow valves; system computer for data analysis and control.
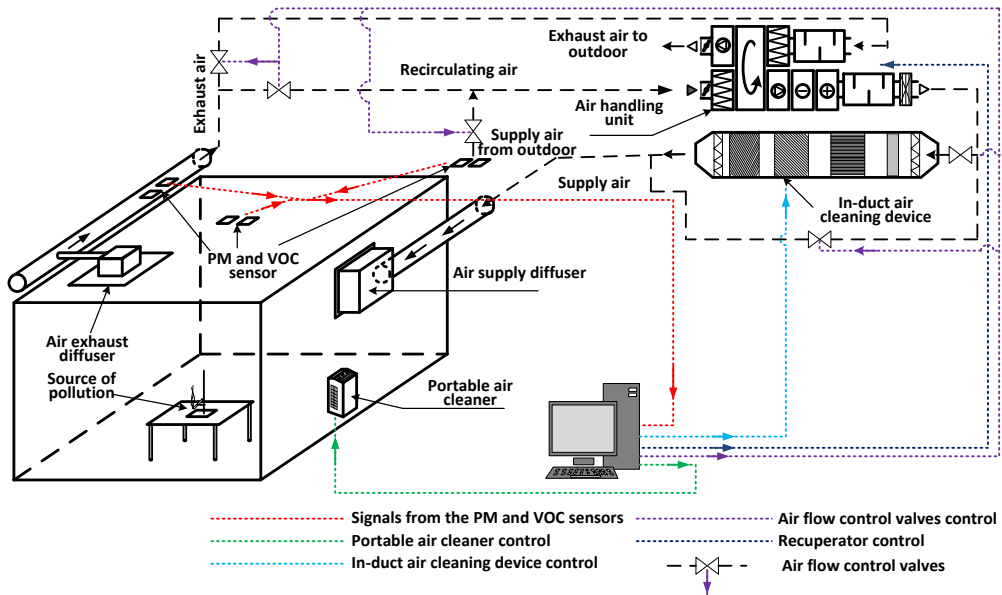
Figure 4.1. Recommended IAQ management scheme.

Generally, this system represents a ventilation based IAQ management system, equipped with advanced management by the signals of the VOC and PM sensors, as well as amended by air cleaning. The latter may be used either in the duct of the supply air or in the room by a portable air cleaner. All of these devices must be connected to the central computer, which applies an algorithm for activating one or several devices based on the real-time information from sensors.

The positioning of the sensors in the centre of the ceiling is more advantageous in the case when IAQ is controlled in several rooms at the same time by the automatic air valves. In such case, it is possible to control IAQ is separate rooms at the same time, providing an opportunity to reduce mechanical ventilation in a room with no air pollution.

It is recommended to use PM and VOC sensors to measure the parameters of the supply air from the outdoors. The air from outdoors, depending on the pollution level, can by treated in the cleaning device or supplied directly. Depending on the quality of the outdoor air, indoor air can also be cleaned and recirculated back to the rooms.

5.   CONCLUSIONS

1.   The indoor environment was investigated in 11 newly built low energy residential buildings. Despite the low air change rate in most buildings (0.08-0.69 h^{-1}), CO_2 and many tested VOC and SVOC concentrations were at reasonable levels, while the concentration of formaldehyde was elevated above the national limit values (3.3-52.3 $\mu g/m^3$). In several buildings, extremely high concentrations of VOCs were observed where the interior installation activities were done just prior to the measurement campaign. A decrease of BTEX concentrations was rapid and fell below the limit values in one month.

2.   The variation of PM and VOC concentrations was assessed for 20 indoor activities, including cooking related sources, other thermal sources, personal care and household products. In most of the pollution episodes, the maximum concentration of particles and VOC in the exhaust air was reached within a few minutes. The most rapid increase in particle concentration (the maximum UFP was measured after 1-2 min after the pollution episode was generated) was during thermal source episodes such as candle, cigarette, incense stick burning and cooking related sources, while the slowest (7-25 min) rise of concentrations was associated with sources emitting ultrafine particle precursors, such as furniture polisher spraying, floor wet mopping with detergent, etc. Placement of the particle sensors in the ventilation exhaust vs. in the centre of the ceiling yielded comparable results for both measured maximum concentrations and temporal variations, indicating that both locations were suitable for the placement of sensors for the management of IAQ.

3.   A combination of ventilation and air cleaning regimes were investigated for the removal of aerosol particles and VOCs in a test chamber, representing a typical room. Air cleaners were effective in the removal of particles, reaching up to 97% removal efficiency based on particle number concentrations after 30 minutes, while $CADR_{PNC}$ varied from 37±4 to 237±11 m^3h^{-1}. The removal of VOCs varied from 21.4 to 45.7% and $CADR_{VOC}$ ranged from 2.2±0.3 to 29.9±2.8 m^3h^{-1}, indicating substantially lower efficiency. The combination of ventilation and air cleaning provided different responses with respect to pollutant removal and energy efficiency. Air cleaning was the most efficient for removing PM from indoor air, minimising the requirement for ventilation. On the other hand, ventilation seemed to be more efficient in the removal of VOCs, while the combination of ventilation and air cleaning increased pollutant removal efficiency by 20% and maximised the energy efficiency.

4.   The development of the model used a grey box approach where the initial data on pollutant variation was collected during the experimental phase, and further applied to the pollutant mass balance model. In the case of incense

smouldering (mostly releases fine particles) with a ventilation rate of 4 h^{-1}, the threshold value is reached after 25 minutes from the start of the pollution episode. If the ventilation rate was kept as at 1 h^{-1}, the threshold value would be reached in 105 minutes. The proposed management approach can be applied to control IAQ in homes, assuring optimal utilisation of the air handling unit in order to achieve the acceptable IAQ in the lowest time span and optimal energy use.

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1. **Čiužas, Darius**; Prasauskas, Tadas; Krugly, Edvinas; Sidaravičiūtė, Rūta; Jurelionis, Andrius; Šeduikytė, Lina; Kaunelienė, Violeta; Wierzbicka, Aneta; Martuzevičius, Dainius. Characterization of indoor aerosol temporal variations for the real-time management of indoor air quality // Atmospheric environment. Oxford: Pergamon-Elsevier Science. ISSN 1352-2310. 2015, vol. 118, p. 107-117. [Science Citation Index Expanded (Web of Science); Academic Search Complete; Academic Search Premier; BIOSIS Previews; Chemical Abstracts (CAplus); Compendex; Science Direct]. [IF: 3,281, AIF: 2,929 (E, 2014)].

2. **Čiužas, Darius**; Prasauskas, Tadas; Krugly, Edvinas; Jurelionis, Andrius; Šeduikytė, Lina; Martuzevičius, Dainius. Indoor air quality management by combined ventilation and air cleaning: an experimental study // Aerosol and Air Quality Research. 2016, DOI: 10.4209/aaqr.2015.10.0577.

3. Mikuckas, Antanas; **Čiuzas, Darius**; Prasauskas, Tadas; Mikuckiene, Irena; Lukas, Romas; Kazanavicius, Egidijus; Jurelionis, Andrius; Martuzevicius, Dainius. A Grey Model Approach to Indoor Air Quality Management in Rooms Based on Real-Time Sensing of Particles and Volatile Organic Compounds // Applied Mathematical Modelling, accepted.

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2. Krugly, Edvinas; **Čiužas, Darius**; Prasauskas, Tadas; Kaunelienė, Violeta; Šeduikytė, Lina; Jurelionis, Andrius; Gagytė, Laura; Martuzevičius, Dainius. Management of indoor air quality by in-room air cleaners and ventilation // WSB 2014: World Sustainable Building Conference [elektroninis išteklius], Management of Indoor Air Quality by In-Room Air Cleaners and Ventilation, 28-30 October, 2014, Barcelona, Spain. Barcelona: Green Building Council Espana, 2014, ISBN 9788469718155. p. [1-6].

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4. Martuzevičius, Dainius; **Čiužas**, **Darius**; Sidaravičiūtė, Rūta; Prasauskas, Tadas; Krugly, Edvinas; Stankevičius, Kazimieras; Kaunelienė, Violeta; Šeduikytė, Lina; Jurelionis, Andrius. Dynamics of aerosol emissions from common indoor pollution sources as characterised in the ventilation exhaust // Aerosols 2014: 3rd workplace and indoor aerosols conference, 13th-16th May, 2014, Wroclaw, Poland: program and abstarcts / Institute of Immunology and Experimental Therapy Polish Academy of Sciences, Wroclaw Branch of the Polish Academy of Sciences. Wrocław: Institute of Immunology and Experimental Therapy Polish Academy of Sciences, 2014. p. 50-51.

5. Martuzevičius, Dainius; Prasauskas, Tadas; **Čiužas, Darius**; Krugly, Edvinas; Sidaravičiūtė, Rūta; Šeduikytė, Lina; Jurelionis, Andrius; Kaunelienė, Violeta. The effect of a sampling location within a room on the dynamics of aerosol concentrations for continuous monitoring of IAQ // IAC 2014: International Aerosol Conference, August 28 - September 2, 2014, Bexco, Busan, Korea. [S.l.]: IAC, 2014. p. [1].

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2. Jurelionis, Andrius; Gagytė, Laura; Šeduikytė, Lina; Prasauskas, Tadas; **Čiužas, Darius**; Martuzevičius, Dainius. Combined air heating and ventilation increases risk of personal exposure to airborne pollutants released at the floor level // Energy and buildings. Lausanne: Elsevier. ISSN 0378-7788. 2016, vol. 116, p. 263-273. [Science Citation Index Expanded (Web of Science); Science Direct; Academic Search Complete; Academic Search Elite; Academic Search Premier; Academic Search Research & Development; Environment Complete]. [IF: 2,884, AIF: 2,447 (E, 2014)].

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5. Prasauskas, Tadas; Martuzevičius, Dainius; Krugly, Edvinas; **Čiužas, Darius**; Stasiulaitienė, Inga; Sidaravičiūtė, Rūta; Kaunelienė, Violeta; Šeduikytė, Lina; Jurelionis, Andrius; Haverinen-Shaughnessy, Ulla. Spatial and temporal variations of particulate matter concentrations in multifamily apartment buildings // Building and Environment. Oxford: Pergamon-Elsevier Science. ISSN 0360-1323. 2014, Vol. 76, p. 10-17. [Science Citation Index Expanded (Web of Science)]. [IF: 3,341, AIF: 2,304 (E, 2014)].

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LIST OF ANNEXES

ANNEX 1. Dynamics of particle size distribution during: cooking related sources; electric stove operating alone (a), water boiling on electric stove (b), cooking oil heating on electric stove (c), Teflon pan heating on electric stove (d); other thermal sources; cigarette burning (e), paraffin (f) and aromatic (g) candle burning, incense smouldering (h); use of personal care products; air-freshener spraying (i), hair dryer operating, medium hot air (j) and hottest hot air (k); Household maintenance sources; furniture polisher spraying (l), floor wet mopping with detergent (m), floor vacuuming (n), electric radiator operating (o), electric air heater operating (p), ironing with steam (r) and without steam (s) operations (dN/dlogDp, p/cm^3).
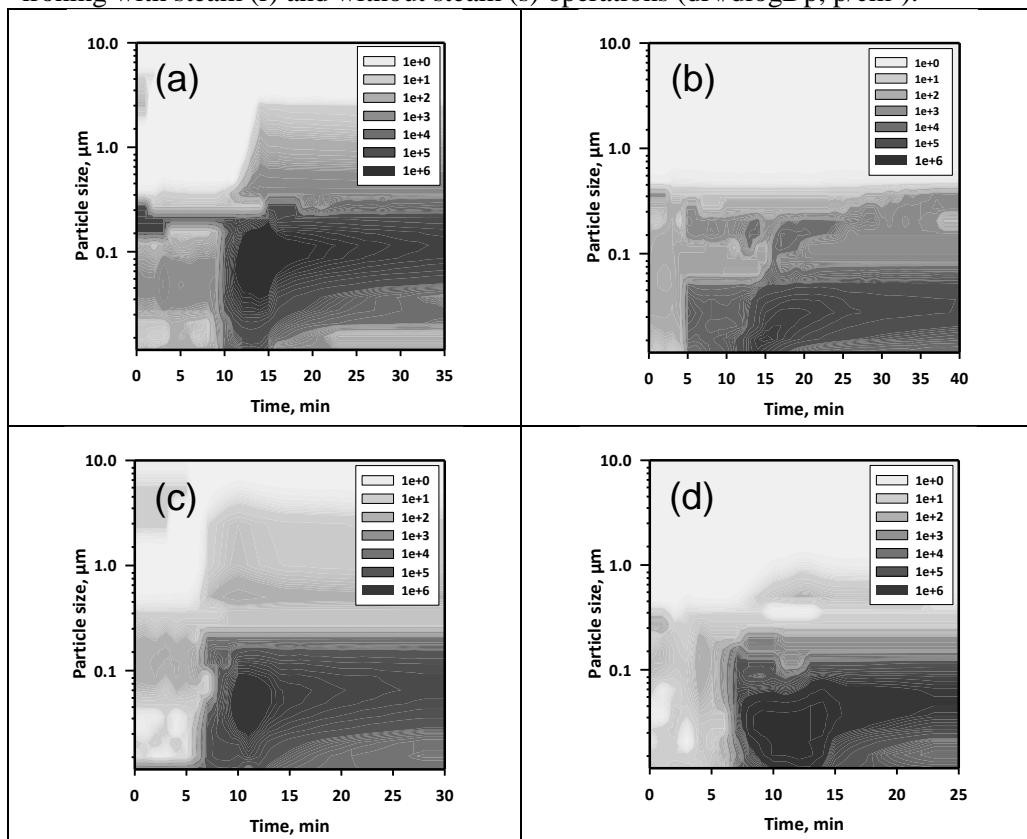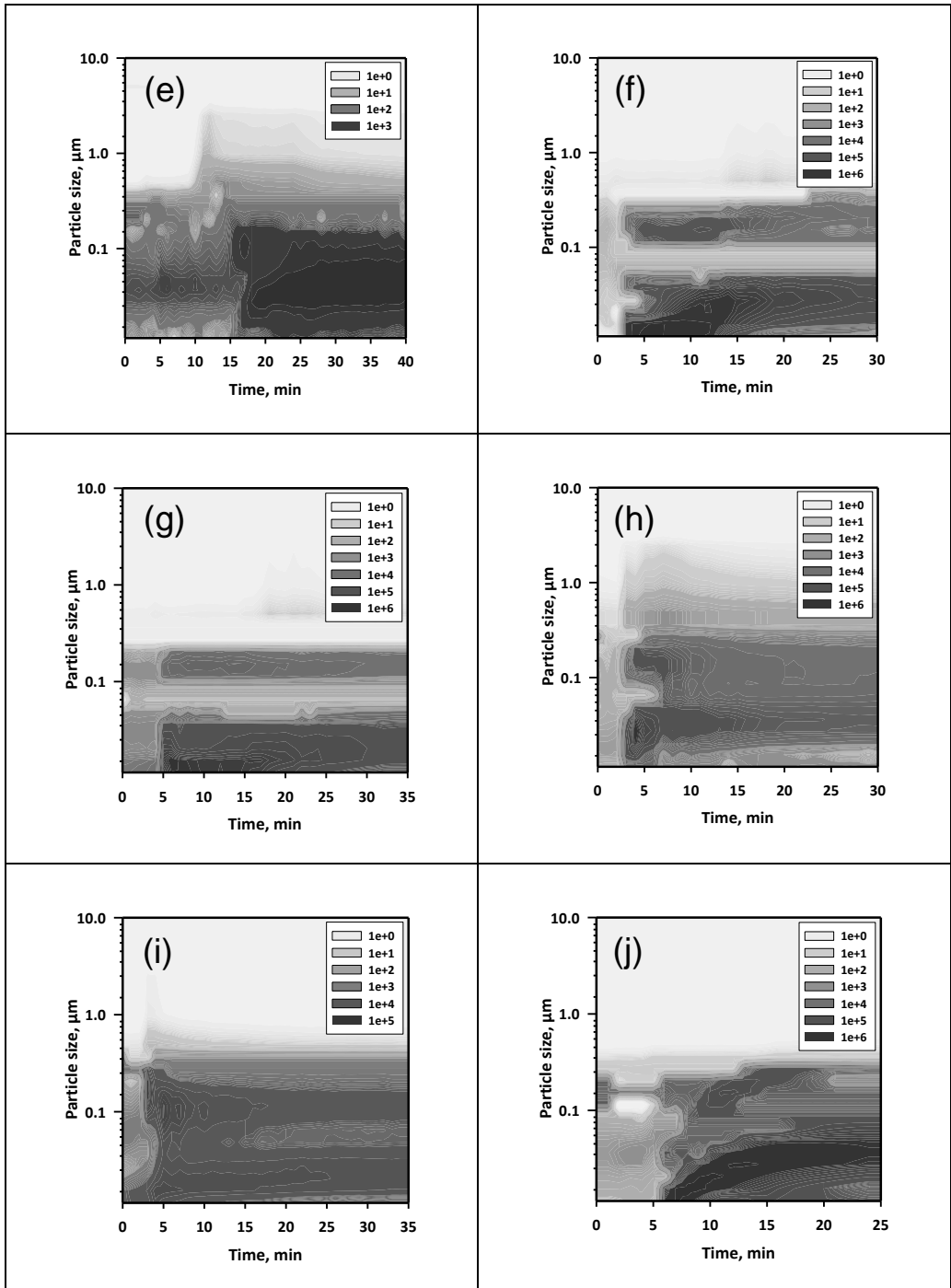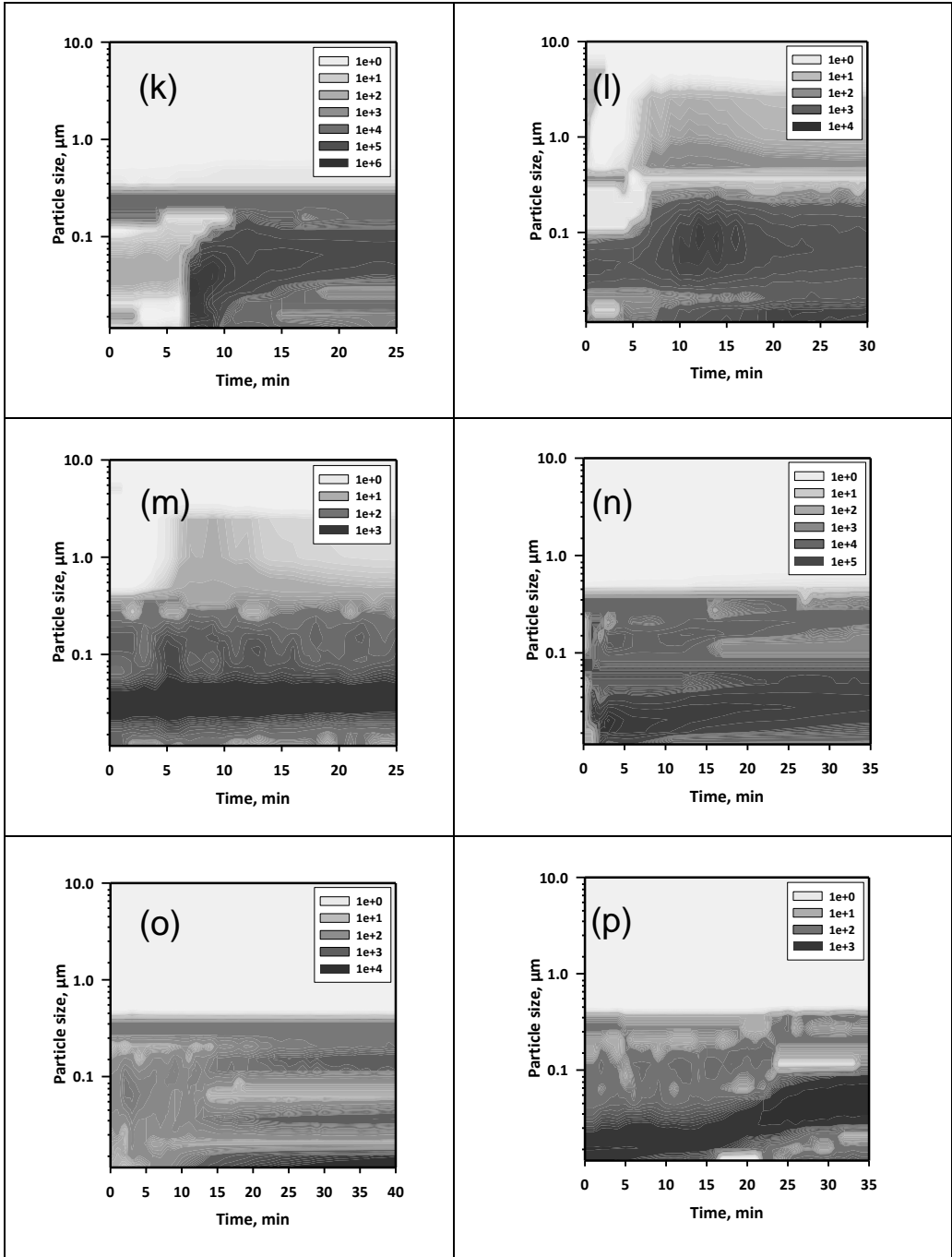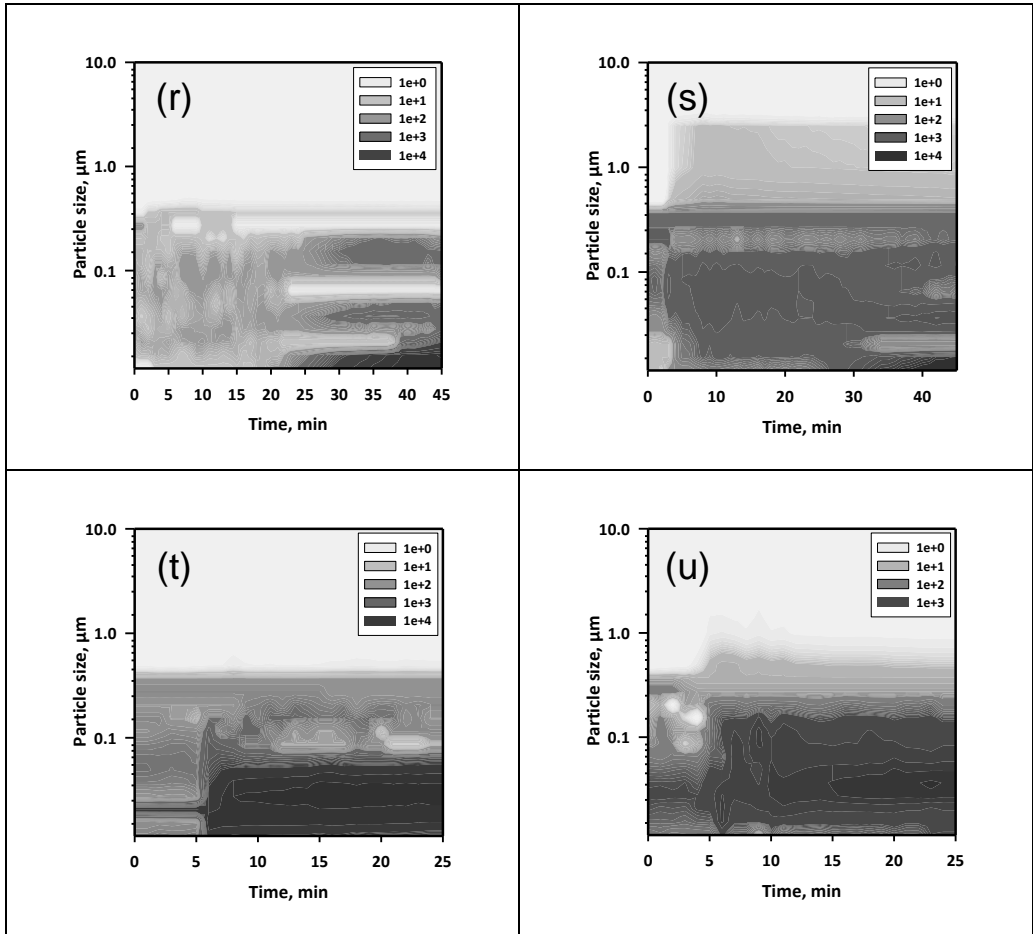
ANNEX 2. CFD prediction results of $PM_{0.5}$ particle concentration isosurfaces in the case of warm contaminant injection (A) and isothermal contaminant injection (B).
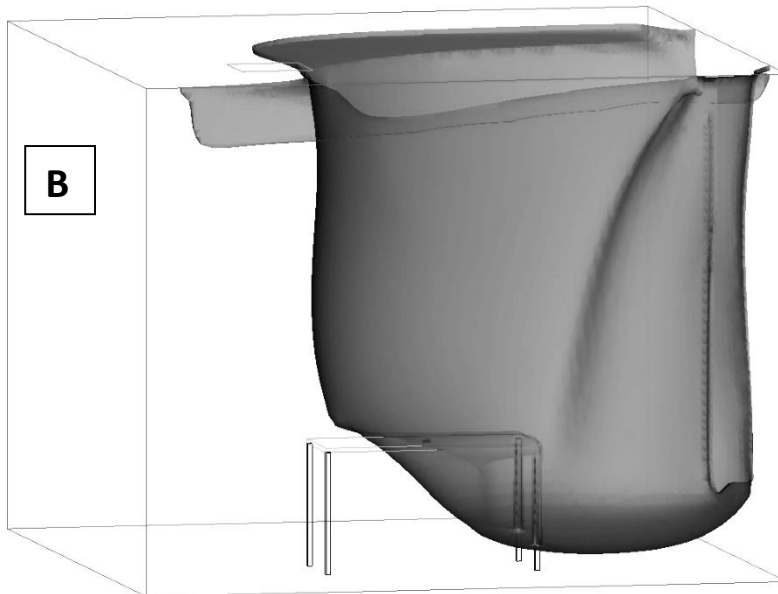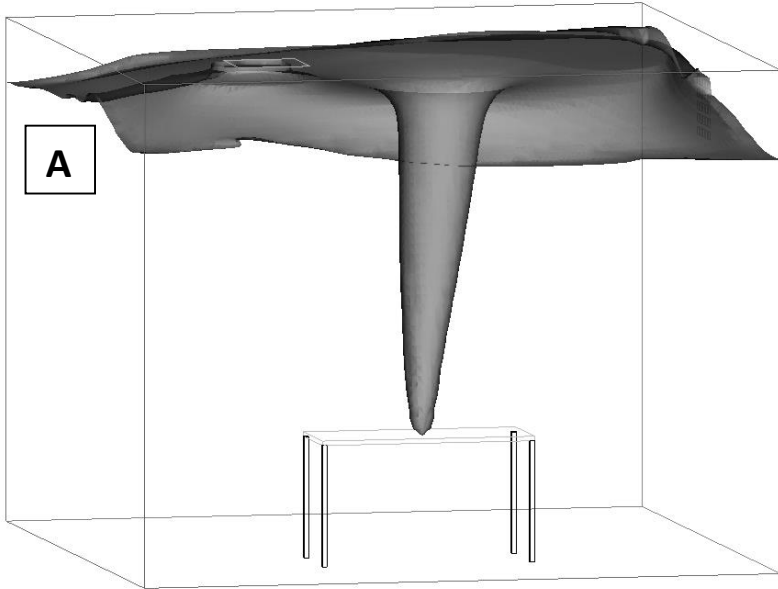
ANNEX 3. Results of different ventilation rates and portable air cleaner C2 operating mode: A – E_{PNC} (%); B – $CADR_{PNC}$ (m^3h^{-1}); C – EEI_{PNC} ($m^3h^{-1}W^{-1}$); D – E_{VOC} (%); E – $CADR_{VOC}$ (m^3h^{-1}); F – EEI_{VOC} ($m^3h^{-1}W^{-1}$).
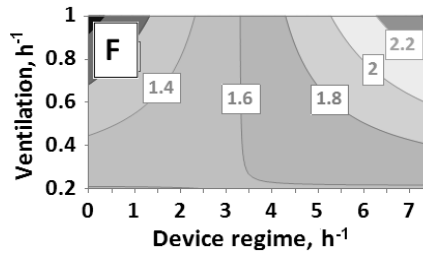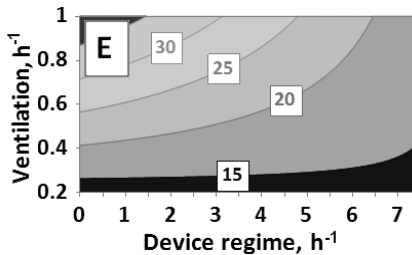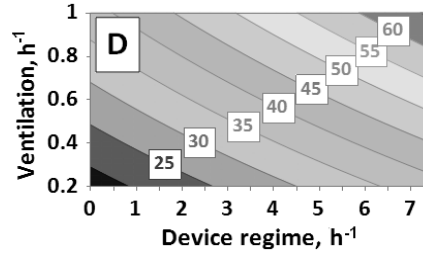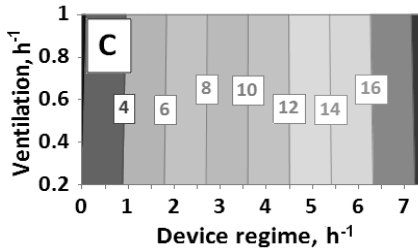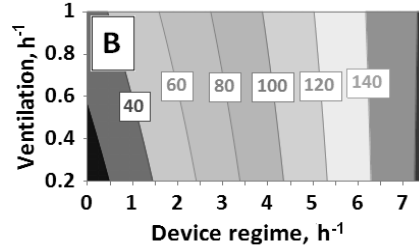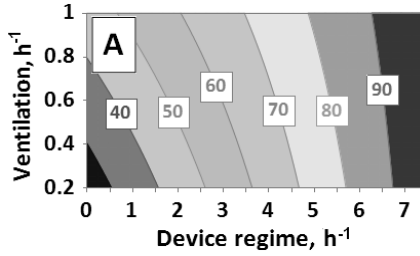
ANNEX 4. Results of different ventilation rates and portable air cleaner C3 operating mode: A – E_{PNC} (%); B – $CADR_{PNC}$ (m^3h^{-1}); C – EEI_{PNC} ($m^3h^{-1}W^{-1}$); D – E_{VOC} (%); E – $CADR_{VOC}$ (m^3h^{-1}); F – EEI_{VOC} ($m^3h^{-1}W^{-1}$).
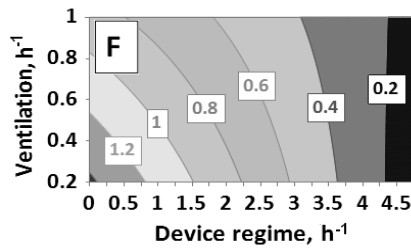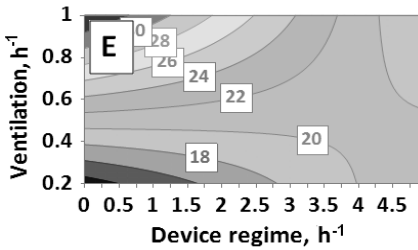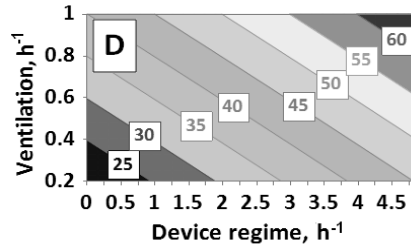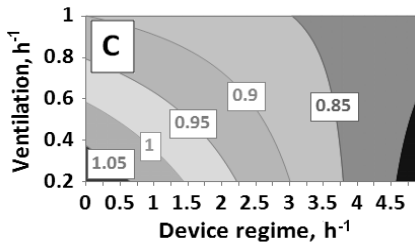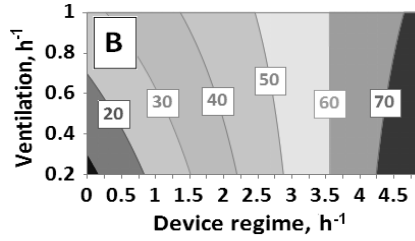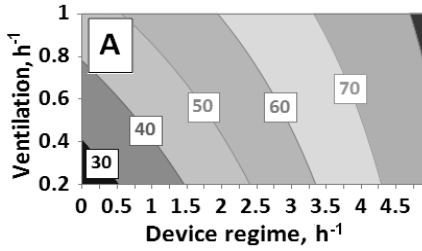
ANNEX 5. Modelling algorithms: A) variation of pollutant concentration in room; B) variation of pollution source; C) model adjusted for particle losses due to deposition.
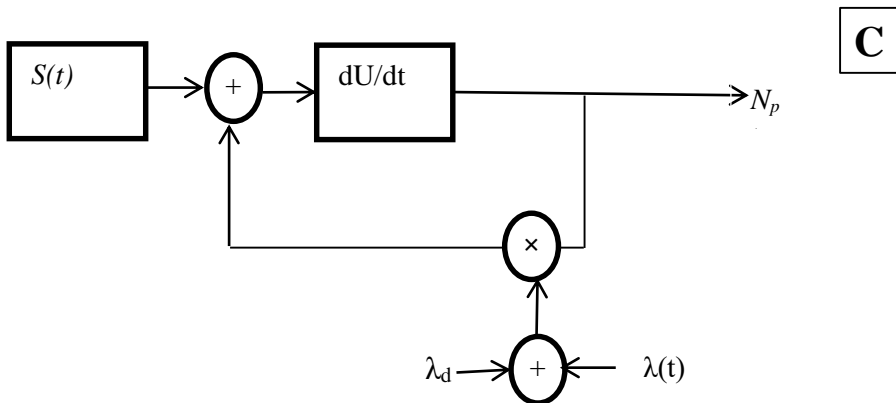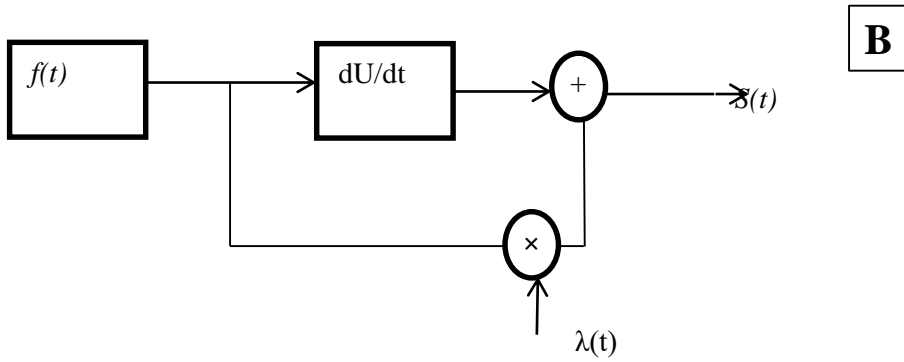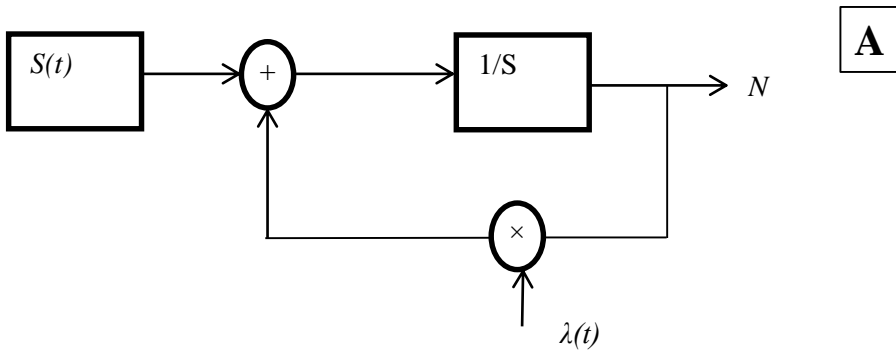
ANNEX 6. Variation of pollutant concentration during onion frying, when ventilation rate is managed excluding fine particle measurements.