KAUNAS UNIVERSITY OF TECHNOLOGY

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# AIR CONTAMINATION BY PARTICULATE MATTER FROM PROCESSES OF BUILDING REFURBISHMENT AND OPERATION

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KAUNO TECHNOLOGIJOS UNIVERSITETAS

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# ORO TARŠA AEROZOLIO DALELĖMIS PASTATŲ ATNAUJINIMO IR EKSPLOATACIJOS METU

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ACH – Air change rate;

APS – Aerodynamic particle spectrometer;

CFD – Computational fluid dynamics;

CV – Coefficient of variation;

EE – Energy efficiency;

EF – Emission factor;

EMEP – European monitoring and evaluation programme;

EPA – Environmental protection agency;

EPBD – Energy performance of buildings directive;

EPC – Energy performance certificate;

EU – European Union;

HEPA – High efficiency particulate air filter;

HVAC – Heating, ventilation, and air conditioning;

GHG – Greenhouse gas;

IAQ – Indoor air quality;

I/O – Indoor/Outdoor ratio;

IEQ – Indoor environmental quality;

ISO – International organization for standardization;

MCE – Mixed cellulose ester;

MMMFs – Man–made mineral fibres;

MMVFs – Man–made vitreous fibres;

OPC – Optical particle counter;

PCOM – Phase contrast optical microscopy;

PM – Particulate matter;

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

PM10 – Particles with a diameter smaller than 10 μm;

PMC – Particle mass concentration;

PMtotal – Total particulate matter;

PNC – Particle number concentration;

PSD – Particle size distribution;

RHc – Relative humidity in cold spot;

RHw – Relative humidity in warm spot;

QA/QC – Quality assurance / quality control;

RH – Relative humidity;

SEM – Scanning electron microscopy;

SD – Standard deviation;

SVF – Synthetic vitreous fibres;

Tc – Temperature in the cold spot;

TEM – Transmission electron microscopy;

To – Ambient air temperature (outdoor);

Tw – Temperature in warm spot;

TWh – Terawatt–hour;

VOC – Volatile organic compounds;

WHO – World health organization.

# INTRODUCTION

While it is widely known that the living environment greatly affects human performance, however little is done to examine the extent of this impact so far (WHO 2011). People spend 60–90 % of their life indoors – at home or in other public or private indoor environments, such as schools, cafes and restaurants (Jantunen 2011; Klepeis et al. 2001; Leech et al. 2002; Yu et al. 2009). Clean air in the living environment is very important for the public health. Indoor air quality (IAQ) is particularly important for vulnerable groups of population, such as infants, children, and elderly people who are suffering respiratory or allergic diseases (Jantunen 2011). The health effects of different indoor pollutants, their concentrations and their public health significance are being studied worldwide. Nowadays is performed a number of scientific studies that emphasize the significant effects of indoor air pollutants on the human health (Crump et al. 2009; Oliveira Fernandes et al. 2008; WHO 2011). Various indoor air pollutants are responsible for exacerbate respiratory diseases, allergies, intoxication and certain types of cancer (e.g. asbestos, radon, environmental tobacco smoke, combustion products, volatile organic compounds, biological pollutants, etc.) (Bayram and Bakan 2014; Henderson et al. 2014; Petry et al. 2014).

IAQ is largely dependent on the outdoor air quality, due to the continuous indoor air mixing with the ambient air (Bernstein et al. 2008; Chen et al. 2011; Habre et al. 2014; Massey et al. 2009; Ruckerl et al. 2011). However, air pollutants in buildings are strongly linked to building indoor factors as well (emissions from building structure, fabrics, coating, furnishing, ventilation system, food preparation, occupant activities, etc.) (Gunschera et al. 2013; Gustafsson 2007; Mendell et al. 2006; Nicole 2014).

Energy Performance of Buildings Directive (EPBD) recast aims to develop Energy Performance Certificate (EPC) to become a real, active energy label of houses. Building renovation is mostly based on economic aspects – performing cost-effective refurbishment actions assuming further savings in energy costs, without taking into account possible changes in air quality conditions. Therefore, a natural question arises whether it will be possible to ensure a comfortable and healthy living for occupants; maybe it is useful to include the assessment of IAQ together with the building modernization and energy audit.

There is a lack of methodologically robust intervention studies that support the improvement of energy efficiency (EE) measures by means of improved IAQ. Along with demonstrating the effects of improving EE on IAQ and health, it is possible to find the most relevant ways to optimize building energy performance that will protect and promote the public health as well as the environment. It is also important to improve the database in order to support the implementation of the related policies in Europe. In this context, overview of the results from before and after intervention measurements in multifamily buildings and emissions from building materials will be presented, and the implications on IAQ will be discussed.

**Aim of the thesis**

To characterize the contamination of indoor air by particulate matter during refurbishment and operation phases of residential buildings.

**Objectives of the thesis**

1. To determine emission factors of size-segregated particulate matter from selected powdery construction materials;

2. To compare emissions of size-segregated particulate matter and mineral fibre from building materials during various mechanical processing operations;

3. To assess mineral fibre emissions from thermal insulation materials, their pathways into the living environment, and removal from indoor environment during building operation phase;

4. To evaluate temporal and spatial variations of aerosol concentrations in living environment in multifamily apartment buildings before and after refurbishment;

5. To develop a set of indicators for researching of factors influencing indoor air quality, based on the spatial and temporal variation of particulate matter;

6. To develop recommendations for the refurbishment and operation of multifamily buildings assuring healthy indoor air quality.

**Scientific novelty**

1. The emission factors for the size-segregated aerosol from building materials aerosolized by fluidization and gravitation methods focusing on the fine aerosol fraction were obtained for the first time;

2. The emission of size-segregated aerosol from asbestos and non-asbestos roof slates were tested in a small-scale chamber and compared against several mechanical processing operations;

3. The unique data was obtained on the variation of air quality during multifamily building refurbishment and operation processes, which has allowed developing a set of indicators for the assessment of indoor air quality based on the temporal and spatial variations of size-segregated particulate matter.

**Relevance of the thesis**

1. Indoor air quality is one of the most important factors influencing indoor microclimate and human wellbeing;

2. Airborne particulate matter is considered to be one of the key pollutants due to its complexity and has been associated with various adverse health effects;

3. Building construction, refurbishment, and operation phases feature multiple sources of air pollution, many of them emitting particulate matter;

4. It is important to identify the possible aerosol particle pathways indoors and to control the exposure of such pollutants during building operation and refurbishment phases.

**Structure and outline of the dissertation**

This dissertation is divided into the following parts: list of abbreviations, introduction, literature review, measurements and methodology, results and discussion, conclusions, recommendations, list of 199 references, list of publications on the dissertation topic and list of annexes. The literature survey and results of the research are presented in 107 pages, including 15 tables and 33 figures.

**Publication of the research results**

The results of this research are presented in two publications, corresponding to the list of the Institute of Science Information (ISI) database, one publication referred in international databases, and reported at 9 international conferences.

**Practical value of the work**

1. The results of the research provide data for the technical and administrative measures for the improvement of indoor air quality in residential buildings;

2. A set of representative indicators for the measurement of particulate matter has been developed based on the obtained results from the indoor air quality assessment. This set of indicators can then be used in future studies and refurbishment projects of different scales;

3. Potential pollution sources of indoor environment, associated with particulate matter were identified and recommendations were provided in order to minimize their negative impact;

4. Experimental set–ups and particulate matter analysis techniques introduced in the study are innovative and can be applied in the similar future scientific studies;

5. The results included in the dissertation are being used within the project INSULAtE (Improving energy efficiency of housing stock: impacts on indoor environmental quality and public health in Europe), co–financed by the EU Life+ -programme.

# 1. LITERATURE REVIEW

## 1.1. Impact of indoor and outdoor pollution sources on indoor air quality

The living environment is very important for overall human wellbeing. IAQ is one of the most important factors influencing indoor microclimate. Majority of population spend their highest share of time indoors (~90 % in developed countries) (Frontczak et al. 2012; Yu et al. 2009). Here the most intensive interaction of humans with the air impurities occurs. Although ambient outdoor quality is the best regulated and controlled by governmental institutions, it has lower impact to human health than indoor air, according to the distribution of the exposure duration. Thus, the indoor air quality of living environment is more important factor than outdoor air quality, although very difficult to control.

It is well known 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, indoor concentrations usually exceed outdoor concentrations, for some pollutants even with an indoor/outdoor ratio up to 20 (Oliveira Fernandes et al. 2008).

The indoor air, as opposed to the outdoor air, is relatively easier to characterize, since most pollution sources are known and have been characterized to some extent (Sawant et al. 2004; Cao et al. 2005; Yu et al., 2009). The trend of studies addressing specific hazardous pollutants, such as polycyclic aromatic hydrocarbons, dioxins and various types of aerosols is increasing (Barro et al. 2009; Petry et al. 2014; Ryan and Beaucham 2013). All of these pollutants can cause acute or chronic health effects. These symptoms are a part of phenomenon named as "sick house" syndrome.

Air pollution sources in buildings are closely related to building indoor factors such as emissions from building structures, combustion processes (fireplaces, candles, etc.), fabrics, coatings, furnishings, ventilation systems, cleaning operations with household chemistry and food preparation, etc. The occupant himself and its specific activities are also one of the major indoor air pollution influencing factors.

The detailed representation of active and passive indoor environment pollution sources are presented in **Figure 1.1.** Major sources of indoor air pollutants could be categorized into the following groups:

- Ambient air pollution (fuel combustion, traffic, urban and industrial activities) comes into the building through the ventilation system or by infiltration (building envelop permeability) (Abt and Suh 2000; He et al. 2005; Hussein et al. 2005; Janssens and Hens 2003; Liu and Nazaroff 2001; Morawska et al. 2008; Zwozdziak et al. 2013);
- Building materials and furnishings (adhesives, paints, insulation materials, wall and floor coverings, etc.) (Afshari et al. 2004; Edwards et al. 2005; Gustafsson 1992; Gustafsson 2007; Kim et al. 2007; Uhde and Salthammer 2007);

- Processes that occur within buildings (combustion, heating, ventilation and air conditioning systems, etc.) (Jantunen and Carrer 2007; Mendell et al. 2006; Wargocki et al. 2004; Weschler 2006);
- Occupants themselves and their activities, (tobacco smoking, use of cleaning products, plant and pet drugs, cooking, paper processing such photocopying, etc.) (He et al. 2007; Jensen and Knudsen 2006; Nazaroff and Weschler 2004; Singer et al. 2006; Spruyt et al. 2006; Wang and Morawska 2008;
- Domestic animals (pet dander) (Raja et al. 2010; Wright and Phipatanakul 2014);
- Water and soil (air pollutants coming through water supply, radon and contaminated soils) (Lamonaca et al. 2014; Lopez et al. 2008; Pilkyte et al. 2006; Rydock and Skaret 2002; Zhang and Smith 2003).



**Active pollution sources**

A – Tobacco Smoking    D – Combustion processes (fireplace, candle, incense)

B – Housework activities    E – Transport emissions

C – Cooking    F – Mechanical ventilation

**Passive pollution sources**

1 – Flooring (carpet, vinyl flooring, painted floors, etc.)
2 – Impregnated wood products (impregnated wood construction, panel floor, etc.)
3 – Chipboard furniture
4 – Wall covering (vinyl wallpaper, emulsion paint, etc.)
5 – Thermo insulation materials
6 – Construction adhesives and sealants
7 – Household chemicals (washing and cleaning chemicals, repellents, pesticides, etc.).
8 – Flammable volatile liquids (gasoline, paint thinners, etc.).
9 – Air fresheners
10 – Leisure handicraft tools
11 – Office equipment (laser printer, computer, etc.).
12 – Natural ventilation

**Figure 1.1.** The illustration of active and passive indoor environment pollution sources

The ambient air introduced into the indoor air through the ventilation system or under natural conditions is also an important factor to IAQ and it isn't always easily controlled source. Outdoor air used for the ventilation of the buildings may also be an important source of pollution, which may contain significant amounts of particulate matter (PM), biological particles (microorganisms, pollens, etc.) and various gaseous pollutants (VOC, $NO_x$ and $O_3$). The pressure differences between the building structures, which are the driving force of the air flows, are also an important source of aerosol particles and gaseous pollutants (Chen and Zhao 2008). The level of various pollutant concentrations present in the ambient air provide a baseline for the level of pollutants found indoors.

According to the scientific literature the relationship between indoor and outdoor pollutants (e.g. PM) can be assessed using three different parameters: 1) Indoor–to–Outdoor ratio (I/O ratio) (the ratio between indoor and outdoor particle concentrations); 2) infiltration factor (the equilibrium fraction of ambient particles that penetrates indoors and remains suspended); and 3) penetration factor (the fraction of particles in the infiltration air that passes through the building shell) (Chen et al. 2011; Liu and Nazaroff 2003; Thatcher et al. 2003). The distribution of measured I/O ratios from the numerous worldwide research studies is presented in **Figure 1.2.** The I/O ratios observed in these research studies varied in a wide range (from 0.02 to 31). The differences among the I/O ratios are attributable to the different indoor particle sources, structure of buildings (geometry of the cracks), outdoor pollution patterns, different ventilation conditions and other factors.
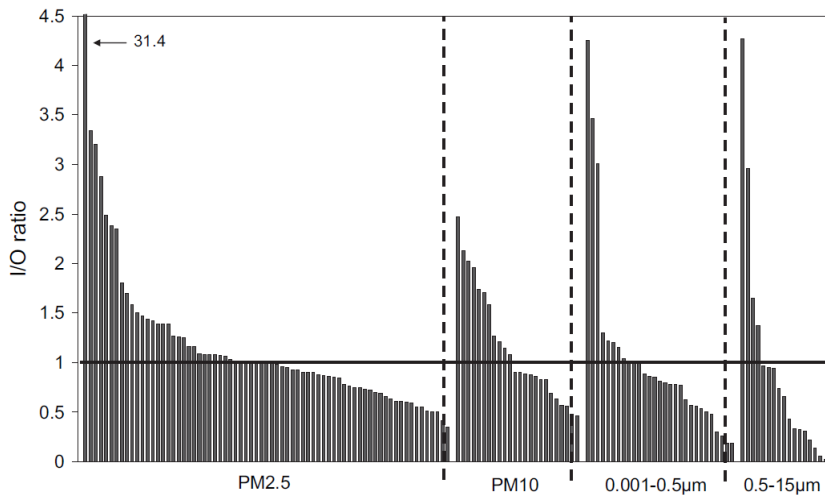


**Figure 1.2.** The distribution of measured I/O ratios from the numerous worldwide research studies (Chen et al. 2011)

Among above described possible aerosol particle pathways to indoor environment, outdoor airborne particles can be brought into indoor environments by humans as well. Outdoor particles can migrate to indoor environments via soil adhering to footwear and then resuspended into the indoor air (Layton and Beamer, 2009), and finally affect the human health.

## 1.2. Health effects related to indoor air pollution

### 1.2.1. Particulate matter – harmful indoor air pollutant

Exposure to PM from household and ambient sources had the most varied pattern with respect to the epidemiological transition. This is partly due to the fact that the exposure and health effects of aerosol particles are heterogeneous (Lim et al. 2012). Household air pollution from solid fuels and ambient PM pollution contributed more than 4 % and 3 % of the annual disability–adjusted life years lost in the 2010 Global Burden of Disease comparative risk assessment (in 1990 – 7 % and 3.5 %, respectively for solid fuels and ambient PM pollution) (**Figure 1.3**). Household air pollution from solid fuels and ambient PM pollution are on 3rd and 9th positions of 20 leading risk factors in 2010, respectively (Lim et al. 2012).



**Figure 1.3.** Burden of disease attributable to 20 leading risk factors in 1990 (A) and 2010 (B), expressed as a percentage of global disability-adjusted life years (Lim et al. 2012)
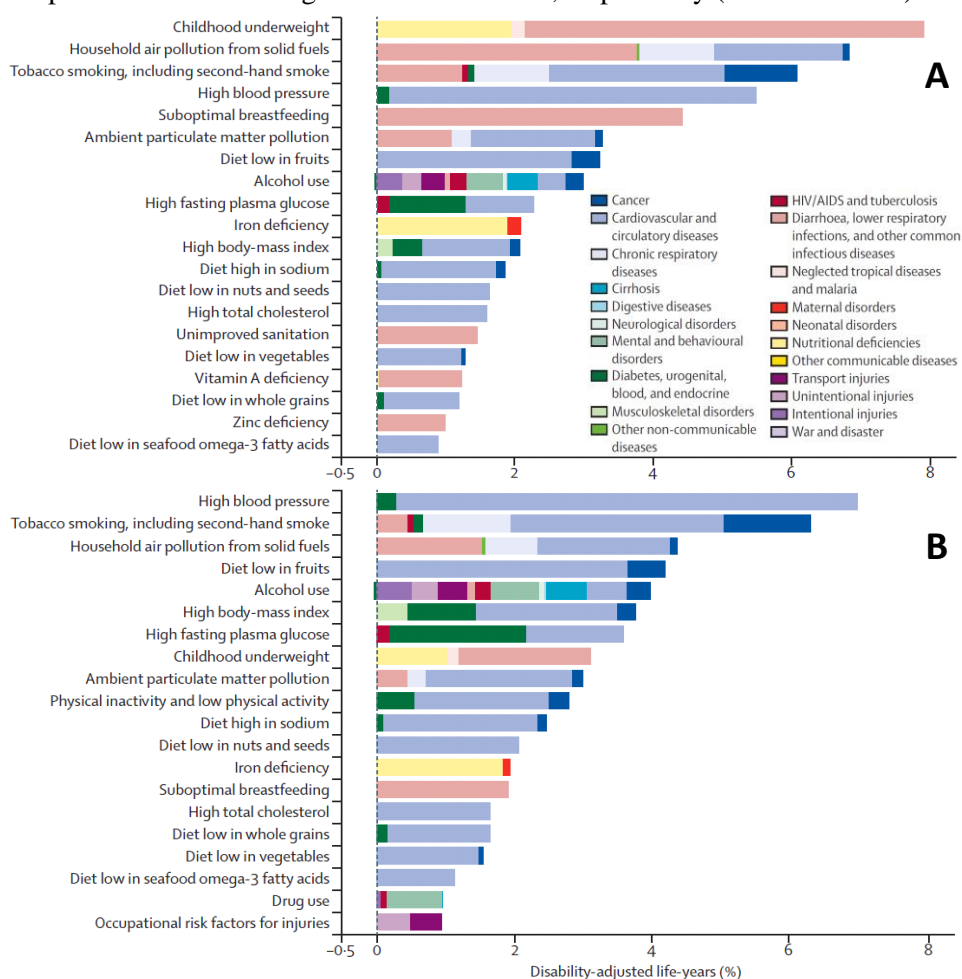
PM in the ambient air has been analyzed in numerous studies for a long period and PM concentrations have been continually monitored for many years. Airborne PM is considered to be one of the key pollutants due to its complexity and adverse health effects. PM, especially its fine fraction, referred to as $PM_{2.5}$ (particles with aerodynamic diameter lower than 2.5 μm) has been associated with various adverse health effects (Pope 2000; Laden et al. 2006; Pope and Dockery 2006; Weichenthal et al. 2007; Dockery 2009; Shimada and Matsuoka 2011). Illustration of compartmental deposition of PM and asbestos fibres in respiratory tract is presented in **Figure 1.4.** $PM_{2.5}$ fraction deposits throughout the respiratory tract, particularly in small airways and alveoli.

There is a substantial and growing evidence that exposures to PM air pollution contributes to the risk of both fatal and non–fatal cardiovascular disease events (Brook et al. 2010; Pope and Dockery 2006). The most consistent findings for an induction of asthma in childhood is related to the exposure to the environmental tobacco smoke, dampness and mould, and living in homes close by busy roads (Heinrich 2011). The particles can contain many different combinations of the hundreds of chemical compounds that have been found to be present in the air in a particle form, thus making them even more harmful.
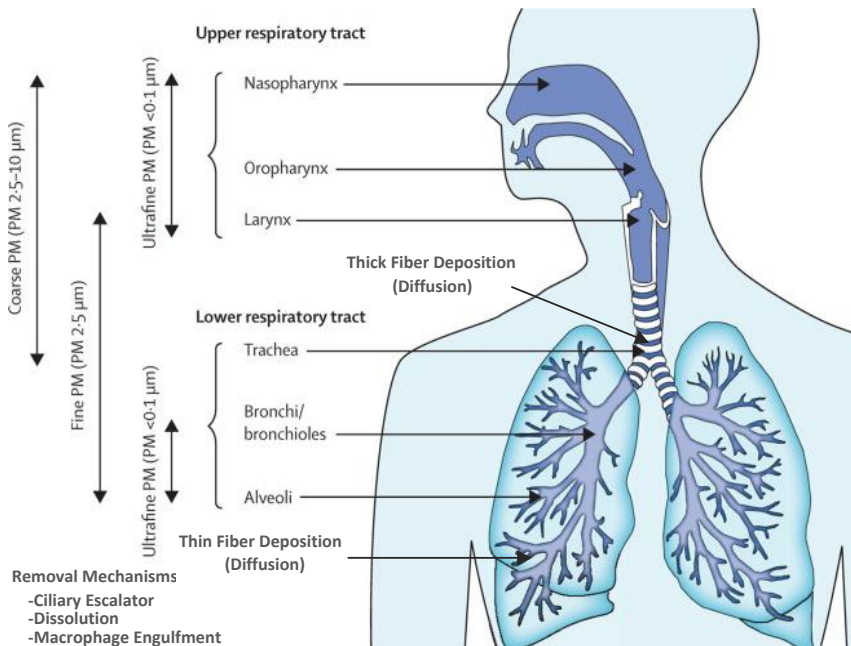


**Figure 1.4.** Compartmental deposition of particulate matter in human respiratory tract (Guarnieri and Balmes 2014; Baron 2001)

A report from WHO (2000) stated that on a global scale, 4 to 8 % of premature deaths are related to the exposure to PM in the ambient and indoor environment. Franck et al. (2011) has revealed significant associations between indoor particle

concentrations and the risks for respiratory diseases in young children, and indicated that short–term measurements can help to assess the health risks of indoor particles. Laden et al. (2006) had concluded that total, cardiovascular, and lung cancer mortality were each positively associated with the ambient $PM_{2.5}$ concentrations while reduced $PM_{2.5}$ concentrations were associated with the reduced mortality risk. Thus, the particle size distribution (PSD) (especially aerodynamic one) of potential hazardous particles is the main parameter controlling their behaviour in the air causing possible health effects.

### 1.2.2  Mineral fibres – harmful indoor air pollutant

Mineral fibres such as asbestos and other man–made vitreous fibres (MMVFs) are part of the Earth's structure. They may enter the environment through natural processes or human activities. Asbestos is a collective name given to minerals that occur naturally as fibre bundles and that possess unusually high tensile strength, flexibility, and chemical and physical durability (Morawska and Salthammer 2003). These attributes of asbestos led to a wide range of industrial applications. At the same time, many types of the MMVFs have been developed for further industrial purposes. It is used primarily for heat and sound insulation of buildings during its construction and refurbishment processes, but they also have numerous filtration, fireproofing, and other applications. Through the miscellaneous use of all these mineral fibres, their infiltration into the environment has steadily increased.

*Asbestos*

Exposure to asbestos during the past decades registered about 20 000 new cases of lung cancer around the world (Tossavainen 1997) and by the year 2050, there will have been ∼90 000 deaths from mesothelioma in the Great Britain (Hodgson et al. 2005). Asbestos is also one of the reasons for 25 800 lung cancer cases registered in Lithuania in male patients during the years of 1965–1994 (Everatt et al. 2007). There is a consistent evidence showing that, for workers exposed to asbestos in the distant past (20–30 years), the risk of mesothelioma is still very high (La Vecchia and Boffetta 2012).

There is abundant epidemiologic evidence showing that asbestos fibres can cause various diseases in humans. Three primary diseases are associated with the asbestos exposure: asbestosis (caused by the inhalation and retention of asbestos fibres), lung cancer, and mesothelioma (an otherwise rare form of cancer associated with the lining around the lungs) (Everatt et al. 2007; Lippman 1990; Bayram and Bakan 2014).

Rapid reconstruction of old building roofs generates considerable quantities of asbestos–cement slate waste which is potentially hazardous to workers and inhabitants' health due to the aerosolization of the asbestos fibres (Baron 2001;

Dodson and Hammar 2006; Harding 2009). Asbestos can be considered equivalent to arsenic and mercury according its impact on health.

A study carried out by Bridle et al. (2006) examined the effect of crushing of asbestos–cement sheets. Sheets were crushed using a digger driven over piles of chrysotile asbestos cement roofing sheets. The authors did not find detectable levels of airborne chrysotile fibres, and claims have been made that the chrysotile asbestos in asbestos cement products is altered, by an unexplained process, into a non-asbestos fibrous material. It was hypothesized that cement replaces chrysotile to less hazardous materials, because small individual particles of cement got attached to the surface of the fibres. Similar allegations have been raised by Deruyterre et al. (1980). This finding was later rejected by Burdett (2006), who has closely examined fibres forming during roofing slate treatment processes and found out that the presence of free fibres in the forming aerosol was significant.

## Man–made vitreous fibres

Mineral fibres are referred as synthetic vitreous fibres (SVF), man–made mineral fibres (MMMF), synthetic mineral fibres (SMF), and man–made vitreous fibres (MMVF), although these terms essentially mean the same product. MMVFs have been commercially manufactured and marketed for the last 60 years. According to the International Agency for Research on Cancer (IARC), in 2001, it was estimated that over nine million tonnes of MMVFs were produced annually in over 100 factories around the world (IARC 2002).

Health concerns of MMVFs are based on the morphological and toxicological similarities between MMVF and asbestos. The hazardous properties of fibres are believed to be attributable to its fibrous nature rather than to its chemical composition. The most common negative health effects associated with MMVFs is temporary skin, eyes, nose, and throat irritation caused by fibres penetrating skin's outer layers (Ziegler–Skylakakis 2004). There is insufficient evidence concerning MMVFs effect on non-cancerous respiratory diseases to the lungs, though the oxidising stress of the cells can also occur, especially in case of repetitive exposure (EASHW 2009). Thus, it is important to characterize and control mineral fibre emissions from construction materials during the refurbishment and operation phase of the building. Fibre toxicity levels can be determined by four key parameters: fibre concentration (fibre/cm$^3$), surface density (fibre/cm$^2$), fibre dimensions (length, aerodynamic diameter, μm), and residence time in the lungs (Vuyst et al. 1995).

## Cellulose fibres

After banning the usage of asbestos–cement sheets (in 2001 in Lithuania), the new generation of cement–based roofing slates has appeared on the market. A typical cement matrix in asbestos cement products consists of calcium hydroxide (10–12 %), calcium silicate hydrates (60–80 %), calcium aluminate hydrates (3–10 %), calcium aluminate sulphate hydrates (0–5 %) and unreacted cement (Burdett

2006). In case of non unreacted asbestos cement, the composition is rather similar, with exception that asbestos fibres have been replaced with cellulose fibres (Shen et al. 2006). Cellulose fibres have been introduced as less hazardous replacement of chrysotile fibres in European Union (EU) (Harrison et al. 1999). On the other hand, there is scientific evidence that these fibres are durable and may cause adverse health effects if inhaled in excessive dosages (Cullen et al. 2002; Warheit et al. 2001; Muhle 1997; Warheit 1998; Cullen et al. 2000). Since cellulose represents a family of materials, there is a great need to assess the toxicity of the various respirable forms of this organic fibre–type (Warheit et al. 2001).

## 1.3. Indoor air quality exposure to particulate matter and mineral fibre emissions

### 1.3.1. Particulate matter emissions during building operation phase

PM, studied in this thesis, varied from nano range (vehicle emissions – 0.01–10 µm; tobacco smoke – 0.01–5 µm) to hundreds of micrometers (asbestos fibres – 0.2–100 µm; MMVF insulation – 2.0–1000 µm). An overview of particle types in the indoor environment and their possible sizes is shown in **Figure 1.5.** The particles can be classified and characterized in many different ways, in the first instance according to their physical, chemical or biological properties. Variation between particles is very high, the smallest and the largest airborne particles can differ in size by up to five orders of magnitude. Airborne particle concentration levels, similar to the size differences, can vary in several orders of magnitude as well.
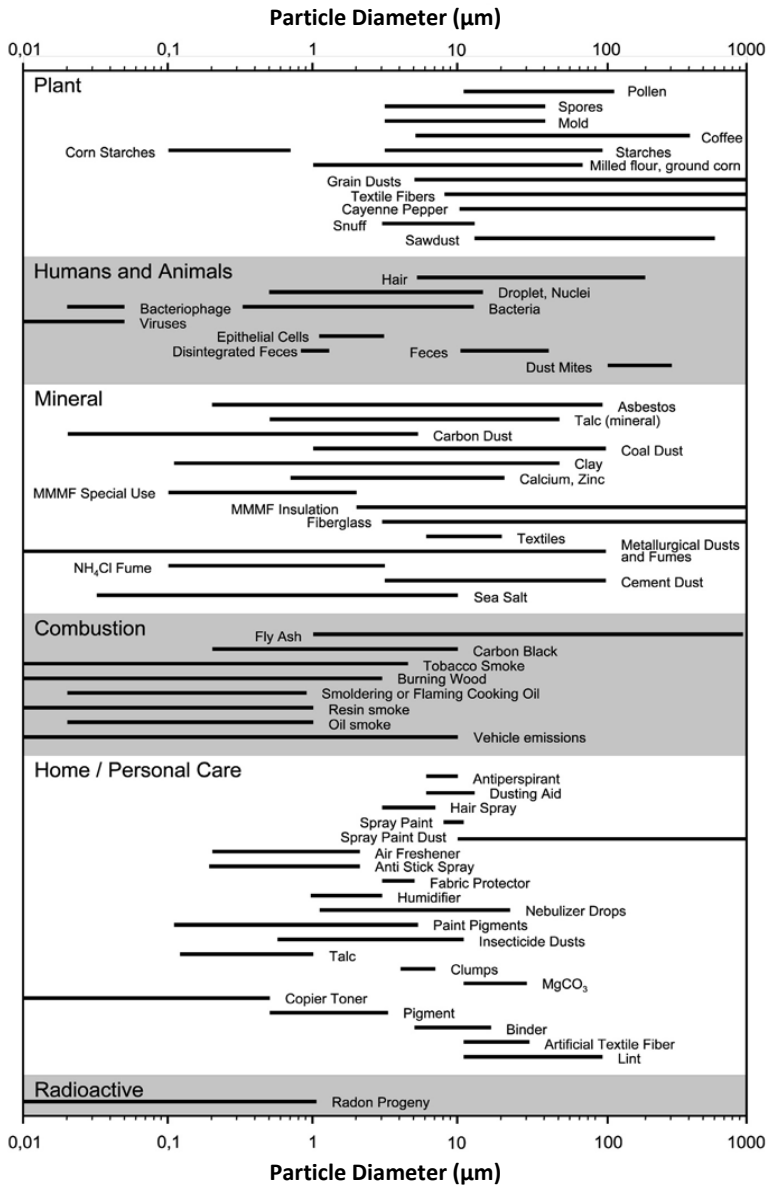
**Figure 1.5.** Size distributions of airborne particles in the indoor environment (Morawska and Salthammer 2003; Owen et al. 1992)

The indoor environment has multiple sources of PM. Among those, outdoor pollution is identified as a key factor influencing indoor air PM concentrations due to its continuous impact (Baxter et al. 2008; Bernstein et al. 2008; Chen et al. 2011; Lai et al. 2004; Massey et al. 2009; Ruckerl et al. 2011; Tippayawong et al. 2009).

Major outdoor particle pathways into the indoor environment are through open windows and infiltration or penetration through cracks and fissures of a building

envelope (Chen and Zhao 2008; Meng et al. 2005). The main urban outdoor PM sources during heating season are fuel combustion, transport exhaust, and atmospheric reactions (Gaydos et al. 2005; Kliucininkas et al. 2011; Lianou 2011).

Multiple sources such as cooking, smoking, candle, and incense burning as well as cleaning activities have been found to be significant PM emitters indoors (Begum et al. 2009; Gao et al. 2013; Klepeis et al. 2003; Nazaroff 2004; Wallace et al. 2006; Hussein et al. 2006; Martuzevicius et al. 2008; Pagels et al. 2009; Guo et al. 2010; Lai et al. 2010; Brown et al. 2012). Type of stove or oven, the time of the year and the floor area in the dwelling affect the concentrations of fine particles indoors as well (Spilak et al. 2014).

During the past decade a number of studies have been conducted involving indoor and outdoor measurements at single–family homes (Massey et al. 2009; Hussein et al. 2006; Pagels et al. 2009; Guo et al. 2010; Lai et al. 2010; Brown et al. 2012; Huang et al. 2007; Puustinen et al. 2007; Chen and Hildemann 2009; Wheeler et al. 2011; Cao et al. 2012; Massey et al. 2012; Fittschen et al. 2013) and multifamily residential buildings (Lee et al. 2002; Stranger et al. 2009; Jung et al. 2010; Wichmann et al. 2010; Diapouli et al. 2011). The most recent studies have focused on continuous measurements of indoor and outdoor ultrafine particle concentrations, including analyses of temporal and spatial variability (Brown et al. 2012; Kearney et al. 2011; Wheeler et al. 2011).


### 1.3.2. Particulate matter emissions during building construction phase

Particle size is one of the most important physical properties of solids which is used in many fields of human activity, such as construction, waste management, metallurgy, fuel fabrication, etc. (Vitez and Travnicek 2010).

The quantification of PM emissions from industrial processes has been mostly characterized by the total PM emissions ($PM_{total}$). The EMEP/CORINAIR Emission Inventory Guidebook (EMEP 2007) has already listed pollutant emissions by size segregated PM, including fractions of $PM_{10}$ and $PM_{2.5}$. However, not all the processes are listed in these inventories. At the same time, the measurement of size segregated aerosol is not compulsory in industrial inventory measurements according to the Lithuanian air pollution legislation. This is only applicable to the ambient air pollution measurements, where the standard for $PM_{10}$ fraction particles (established for protection of human health) for one calendar year is equal to 40 $\mu g/m^3$, and the rate for one day is – 50 $\mu g/m^3$ (it cannot be exceeded more than 35 times per calendar year). The existing limit for $PM_{2.5}$ fraction is equal to 27 $\mu g/m^3$ in the year 2012 (LR aplinkos, 2010). Thus, there is a missing link between emissions and air quality standardization, and it needs to be filled in.

In order to assess the impacts of stationary sources of pollution to the environment and to simulate pollutant dispersion, it is necessary to determine the PM emissions from stationary pollution sources according to the PSD. One of the ways how to measure the dustiness of the bulk material is described in the European

standard (EN–15051 2006). This standard provides details of the design and operation of the continuous drop test method that classifies the dustiness of solid bulk material in terms of health–related fractions.

PM can be produced during the extraction of many construction materials, various industrial processes as well as on a building site. According to Berge (2009) polluting particles may also be a problem during a demolition phase. The so–called material pollution, due to the construction activities, consists of dust (PM) emissions from various powder building materials that are exposed to physical or chemical activities.

The construction aerosol was an object of numerous studies worldwide. Muleski et al. (2005) in their paper had summarized the results of a multi–year research program undertaken by the EPA's National Risk Management Research Laboratory to assess PM emissions from the construction activities. Several studies dealt with the assessment of PSD and emission factors from the various industrial plants e.g. cement production (Ehrlich et al. 2007; Canpolat et al. 2002). Ehrlich et al. (2007) determined the PSD of the fine dust ($PM_{10}$, $PM_{2.5}$ and $PM_1$) in the waste gases from the various industrial plants using eight or six stage cascade impactors (Anderson and Stroehlein impactors) in several regions of Germany. This study has shown that a cascade impactor working under similar sampling conditions is a suitable technique for determining PSD of various dust, only the selection of the sampling period should be taken into account trying to avoid overloading of the impactor stages.

In the ambient air study by Santacatalina et al. (2010) in Southeast Spain excessive levels of $PM_{10}$ and $PM_{2.5}$ were registered in 2006 and 2007, which could be attributed to specific sources, mainly from large public construction work (highway construction), cement and ceramic manufacturing. It was observed that the main sources influencing an ambient $PM_{10}$ fraction are from mineral sources and these pollution episodes were attributable to the public construction work. Recently, the potential hazard of building materials (typically from concrete, cement, wood, stone, and silica) to human health related to the release of particulate matter was described in the wide press by Gray (2010).

### 1.3.3. Mineral fibre emissions during building operation phase

Mineral fibre concentration in settled dust is one of the parameters describing the indoor air quality. This value is expressed by fibre number per area or volume unit (fibre/cm$^2$ or fibre/cm$^3$) depending on the fibre sample collection method. National supervisory authority for welfare and health in Finland has drafted new housing health instructions with new guidelines for asbestos and industrial fibres which will be validated in 2015. Therein provided that "no fibres on surfaces" and "0.2 fibres/cm$^2$ (surfaces)" should be applied for asbestos and mineral fibres, respectively.

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During last decades of previous century, as well as in the first decade of this century, many studies have been performed on worker's exposure and asbestos fibre emissions from various sources, including asbestos–cement sheets, in living areas, commercial and industrial sites (Ansari et al. 2007; Burdett 2006; Campopiano et al. 2009; Gualtieri et al. 2009; Kakooei et al. 2009; Krakowiak et al. 2009; Spurny 1989). Asbestos fibre concentrations during demolition by removal of whole sheets averaged from 0.3 to 0.6 fibre/cm$^3$ for roofs and less than 0.1 fibre/cm$^3$ for walls (Brown 1987). Average values for the concentration of asbestos in the workplace atmosphere were 0.1 fibre/cm$^3$ (Smolianskiene et al. 2005). Surface corrosion was found to be an important factor concerning elevated fibre emissions (Dycze 2004). The release of the fibres during various mechanical operations has been also studied (Burdett 2006; Preat 1993; Pastuszka 2009).

Camilucci et al. (2001) have revealed that materials containing MMVFs generally found in public buildings have a high content of respirable fibres. It is important to ensure high–quality of installation to prevent MMVFs from entering the indoor air. MMVFs can enter the premises via various pathways. Vibrations or carelessly performed refurbishment work can accelerate the appearance of micro-cracks in the walls and the ceilings, which can lead to penetration of mineral fibres through it (Shneider et al. 1990). Mineral fibres from thermal insulation can be carried into the indoor environment via air flow due to the ventilation system irregularities or design errors, as well as through open windows (Kovanen et al. 2007).

## 1.4. Impact of energy efficiency on indoor air quality of residential buildings

### 1.4.1. Future insights on energy efficiency in residential buildings

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 oblige Member States that by 31 December 2020, all new buildings should be nearly zero–energy buildings (Commission 2007). National policies and programs are developed in order to fulfil the EPBD aiming to minimize energy performance for new and renovated buildings (Directive 2003).These requirements relates to the increasing issue of climate change, which in turn is named as the primary threat to public health in 21st century (Costello et al., 2009). The increasing EE in housing sector is one of the most feasible means to reduce emissions of greenhouse gases (GHG). This usually means improving EE and air tightening of the building envelope. Therefore, it is very important what engineering means will be applied to supply suitable microclimate and indoor environment quality (IEQ). This issue is relevant to both newly built and renovated buildings. Without assuring proper means for pollutant abatement and dispersion, a high risk to human health arises in such buildings (Bone et al. 2011).

Measures on improving energy efficiency and decreasing carbon dioxide emissions of buildings sector can be divided into ten principles according to a SQUARE (A System for Quality Assurance when Retrofitting Existing Buildings to Energy Efficient Buildings, SQUARE 2007). Main principles are: improvement of exterior thermal insulation, improving air tightness, optimizing heating and ventilation systems, natural cooling, user's behaviour, etc. All of these can have impact to indoor environment and some common impacts are listed in report (SQUARE 2007), without any measured verification of these impacts. Yet, most of the above mentioned energy measures can be assessed quantitatively by measuring energy consumption, air tightness, air change (ventilation) rate and other factors, which are also required for energy certification (Directive 2003).

In Lithuania, a national program for refurbishment of multifamily buildings was started in 2005; up to 50 % state support could be obtained. As a result, a total of 24,000 buildings were planned to be refurbished with expected energy savings of 1.7 TWh per year (BKA 2007). The current economical situation has caused changes in the implementation of the program, but the overall aim to improve EE of the housing stock remains.

The thermal quality of the building stock has been changed significantly after the collapse of the former Soviet Union, in Lithuania. Since 1992 when the National Building Code was introduced, the required U–values of the building elements are approaching the ones applied in the Scandinavian countries. However, the buildings constructed earlier represent the old style of construction, requiring high energy consumption for heating (Stankevicius et al. 2007). It has been estimated that about 66 % of the Lithuanian population lives in multifamily houses built before 1993. Some 5000 multifamily buildings (majority has been built during the period of the Soviet Union) are located in the Kaunas city, of which 26 % were built before 1960, 65 % between 1960 and 1990, and the remaining 9 % after 1990. The average age of the buildings is about 40 years. These buildings are known for their leaky envelope, low thermal insulation, and unbalanced both heating and natural ventilation systems. The combination of these parameters results in a wide variety of IAQ issues.

In Lithuania, many residential areas rely on combustion of solid fuels as heating sources. The air supplied to ventilation systems is often polluted with products of incomplete combustion (polycyclic aromatic hydrocarbons, dioxins, etc). Thus, even at sufficient air exchange rates in homes, air quality monitoring and control systems are necessary for providing suitable air quality in homes. Yu et al. (2009) emphasized that future research should be directed towards the improvement of health, comfort and energy savings. The integrated air quality management system allows addressing all these priorities.

### 1.4.2. Indoor air quality in refurbished energy efficient buildings

The importance of buildings in environment and health policy is also evident. The fourth ministerial conference on environment and health (WHO 2004) observed

the need for environment and health to be at the core of policies on housing and energy use. The World Health Organization (WHO) resolution on environment and health has called for policies to protect public health from the impacts of major environment–related hazards such as those arising from climate change and housing. The Environment and Health Action Plan for Europe (EP 2005) includes housing among its priorities.

Increasing requirements for the building EE raise new challenges for IAQ management. The main goal of the building refurbishment process is energy saving and improvement of building systems, but the improvement of occupants' wellbeing should also be considered as one of the most important refurbishment goals. From this perspective, IEQ research in low energy/refurbished buildings is of high importance. The modification of building systems, including structures (e.g. insulation of external walls) and heating, ventilation, and air conditioning (HVAC) systems, and new building materials, may have a significant influence on IAQ and subsequently, PM levels.

Recently, WHO Housing and Health Programme implemented a health-monitoring project in Frankfurt, Germany (WHO 2008). The project aimed to assess the potential health impacts of thermal insulation changes in residences. Preliminary results indicated positive impacts on thermal conditions and comfort; renovation and insulation activities did not appear to be in conflict with the health of residents in this climate. However, these results could not necessarily be generalized on a larger scale. It has been estimated that at least some countries, for example Lithuania, are seeing a downward trend in ventilation rates due to the energy conservation concerns. With policies solely focusing on creating more airtight dwellings, exposure to indoor air pollutants may increase, resulting in adverse health effects. An important conclusion from WHO housing and health symposium was that the priority of housing hazards differed "strongly from country to country, as they are influenced by several factors – e.g., cultural, social, economic, building, climatic, geographic"; the effect of housing hazards on health are shared but the prevalence and priority of these hazards differ by country (WHO 2001). Consideration of these priorities is crucial for elucidation of the relevant policies improving living conditions both now and in the distant future.

Overall, there is a lack of methodologically robust intervention studies that support the improvement of EE measures by means of improved health. Moreover, there are almost none large–scale studies on this topic and those that exist are on national scale only. Whereas current national policies regulate improving EE of new buildings, in the future, regulations will be extended for existing (renovated) buildings. There lies a risk that the hydro–thermal behaviour of building structures and IEQ may decrease. Spengler et al. (2011) in his report provided recommendations for Environmental Protection Agency (EPA) to ensure that building weatherization and EE efforts not generate new indoor health issues or worsen existing air quality. Among concerns cited are EE updates (refurbishment) of older buildings, use of untested or risky upgrades, and other alterations that could

generate mould–causing dampness, poor ventilation, excessive temperatures, and finally emissions from building materials that may contribute to health problems.

Increasing insulation of building envelopes can affect IEQ in two ways: 1) improve thermal comfort, and 2) reduce natural ventilation, which in turn may increase concentrations of pollutants in the air. Moreover, various building material emissions may also contribute to decreased air quality. These pollutants may be transported to other zones by ventilation, air conditioning or heating systems and subsequently affect inhabitants. These changes are having important impacts on exposure to indoor air pollutants in residential buildings and present new challenges for professionals interested in assessing the effects of housing on public health (Milner et al. 2011).

Based on the results obtained from this thesis it is planned to comprehensively demonstrate the impacts of building EE on IAQ measurements of airborne PM and mineral fibres from settled dust, i.e. objective and quantitative measures. Along with demonstrating the effects of improving EE on IAQ, the work aims to propose the most relevant possibilities for optimal building energy performance that will protect and promote the public health as well as the environment, and to improve the knowledgebase in order to support the implementation of the related policies in Europe.

## 1.5. The findings of the literature review

1. IAQ is one of the most important factors influencing indoor microclimate. IAQ is mainly affected by strong indoor pollution sources (e.g. combustion processes, food preparation, domestic animals, emissions from furnishings, coatings) and continuous impact from outdoor air. Outdoor air pollution impact to indoor air is associated with the building structure defects (e.g. microcracks) and can be evaluated by I/O ratio, infiltration and penetration factors. If outdoor air does not respect WHO guidelines, air cleaning might be needed (e.g. mechanical ventilation). Pollution source control is recognized as the priority strategy to manage the exposure indoors; therefore its potential should be explored in particular.

2. The summary of the literature review confirmed the negative impact of PM on occupants' health. Airborne PM is considered to be one of the key indoor pollutants due to its complexity and adverse health effects. Fine fraction of PM ($< PM_{2.5}$) has been associated with various fatal and non–fatal cardiovascular diseases. It is crucial to identify and control the levels of PM in the indoor environment with the aim to ensure a healthy living.

3. Indoor air contamination with PM and mineral fibres could be identified as from the building construction inseparable process. Particular air pollutants are generated during different building construction/refurbishment operations (e.g. removal of asbestos–cement sheets, operation with mineral fibre insulation or

powdery building materials). Substantial amount of fine PM are aerosolised into the ambient air during such construction processes, which can affect workers and occupants performance in a negative way. PM and mineral fibres can easily penetrate into the living environment, therefore in order to minimize the environmental impact any preventive actions should be considered.

4.  Nowadays it is evident that energy concerns and climate change are the main factors transforming the overall housing conception and beside these transformations the health outcomes should not be left behind. Without assurance of sufficient ventilation (air exchange rate), application of healthy building materials and appropriate performance of construction works it is impossible to ensure a good IAQ. Only tightening the building envelope and this way reducing natural ventilation will decline the indoor air exfiltration outdoors and increase the residence time of contaminants indoors.

5.  Earlier performed research studies focused only on topics related to building EE. Therefore a more comprehensive analysis on IAQ is essential. It is obvious that effective assessment of interactions between climate change, building environment, EE, and public health demands move from a traditional disciplinary approach to integrate knowledge from all sectors involved. However, complex research studies covering all these factors (EE, environment, and health) are essential not only on a national level but worldwide as well.

## 2. METHODS AND MEASUREMENTS

The methods of the thesis were divided into two major stages: PM/mineral fibre emission measurements in construction/refurbishment phase and assessment of PM/mineral fibre concentrations during building operation phase. Detailed illustration of stages of the doctoral thesis is presented in **Figure 2.1**.

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┌─────────────────────────────────────────────────────────────────┐
│              Construction/Refurbishment phase                     │
└─────────────────────────────────────────────────────────────────┘
┌──────────────────┐   ┌──────────────────────┐   ┌──────────────────┐
│ 2.1.1 Powdery    │   │ 2.1.2 Asbestos and   │   │ 2.1.3 Mineral    │
│ building         │   │ non–asbestos cement  │   │ fibre            │
│ materials        │   │ roof slates          │   │ emissions        │
└──────────────────┘   └──────────────────────┘   └──────────────────┘

┌─────────────────────────────────────────────────────────────────┐
│                      Operation phase                              │
└─────────────────────────────────────────────────────────────────┘
┌──────────────────┐   ┌──────────────────────┐   ┌──────────────────┐
│ 2.1.3 Mineral    │   │ 2.2.1 PM in          │   │ 2.2.2 Comparison │
│ fibre in settled │   │ multifamily          │   │ of PM before and │
│ dust in          │   │ apartment buildings  │   │ after            │
│ residential      │   │                      │   │ refurbishment    │
│ buildings        │   │                      │   │                  │
└──────────────────┘   └──────────────────────┘   └──────────────────┘
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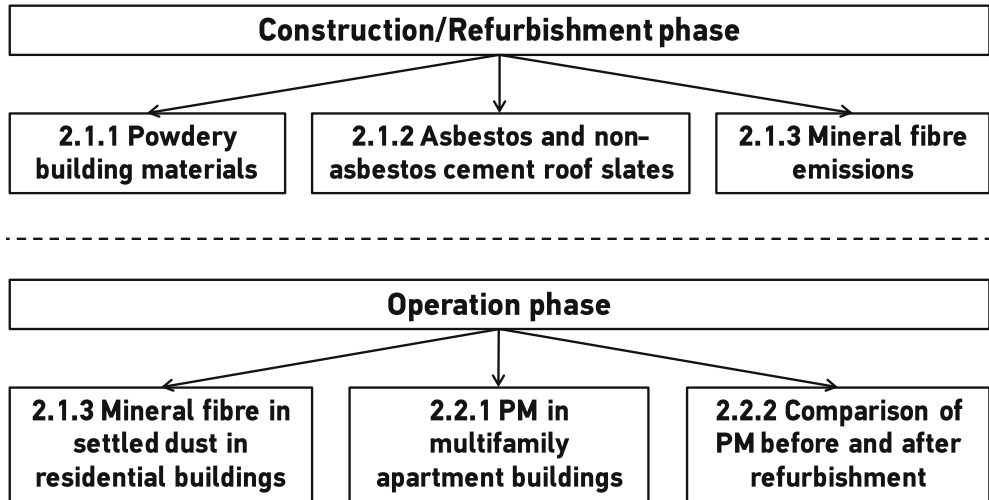
**Figure 2.1.** Stages of the doctoral thesis

### 2.1. Aerosol emissions from building construction and refurbishment processes

### 2.1.1. Particulate matter emissions from powdery building materials

Eleven building materials, commonly used in construction work, were chosen for determination of particle size distribution (in alphabetic order): 1) cement; 2) chalk; 3) clay; 4) gypsum; 5) hydrated lime; 6) masonry grout; 7) quartz sand; 8) sand; 9) structural lime; 10) wood grinding dust and 11) wood sawdust. Building materials used in the laboratory study were selected in accordance to in reality used building materials to ensure as possible uniform PSD, as well as their shape, composition, and other important properties (Kaya et al. 1996).

PSD of various powdery building materials were assessed by two aerosol generation methods: fluidization (dust is resuspended by direct entrainment into airflow in a metal tube) and gravitation (a source sample fell as a discrete slug through the air into an enclosed chamber from which dust is evacuated). These two methods represent actual industrial activities, such as pneumo–transportation, batching and unloading, and are easily modelled under laboratory conditions (Gill et al. 2006).

In a case of fluidization (**Figure 2.2**), 0.1 g of powdery building material was inserted into an injection tube. The material sample was dispersed into the experimental chamber by a short gust of compressed air. The air was withdrawn from the chamber via sampling tubes together with sampled particles. In a case of gravitation (**Figure 2.3**), 1.0 g of powdery building material was dropped from 40 cm height into an enclosed chamber from which dust was evacuated.

In both cases, PSD was determined by an Aerodynamic particle sizer (APS, model 3321, TSI Incorporated, Shoreview, MN, USA) and an optical particle counter (OPC, model 3016IAQ, Lighthouse worldwide solutions, Fremont, CA, USA). Additionally, the PSD based on mass concentrations was determined using a three–stage cascade impactor (PM10 Impactor, Dekati Ltd., Finland). PM samples were collected on an aluminium foil plates whose surfaces were covered with a thin layer of vacuum silicon paste (Dow Corning 732, Dow Corning Corp., USA) to prevent the particle bounce–off. Before starting a new measurement, the inner surface of the chamber was covered with a fresh aluminium foil and was replaced during each new measurement.

The APS sizes particles in the range from 0.37 to 20 micrometers using a sophisticated time–of–flight technique that measures aerodynamic diameter in real time, dividing into 52 channels, with the sampling flow rate of 1.0 l/min. The OPC measures particle concentration in 6 channels: 0.3 – 0.5 μm; 0.5 - 1 μm; 1 – 2.5 μm; 2.5 – 5 μm; 5 – 10 μm; >10 μm, with the flow rate of 2.88 l/min. During impactor measurements, the particles were collected in the fractions of < 1 μm; 1 – 2.5 μm; 2.5 – 10 μm; and >10 μm, with a flow rate of 10 l/min. A three stage cascade impactor was confirmed to be a suitable technique for investigation of powdery building materials, if the total particle mass concentration does not exceed the allowable limit value of 40 mg/m$^3$ as half–hourly averages under standard conditions (273 K, 1013 hPa, dry gas) (ISO–23210 2009).

Aerosol was sampled by separate tubes at the same height (20 cm from the chamber bottom). Tubes from APS and OPC were inserted into different sides of the chamber. The supplied air for particle resuspension was cleaned using a high efficiency particulate filter (HEPA, class 12). The air was withdrawn from the chamber via sampling tubes together with sampled particles.

Aluminium substrates (used with a three stage cascade impactor) were weighted twice before and after the sampling by a microbalance with sensitivity of ±1.0 μg (model MX5, Radwag, Poland). The substrates were equilibrated in a thermostat at the room temperature at relative humidity of 40–50 % for 24 hours. Laboratory blanks were collected and analyzed for $PM_{10}$ and $PM_{2.5}$ fractions to reduce gravimetric bias due to the filter handling during and/or after sampling. From the weight differences and airflow rate $PM_{10}$ and $PM_{2.5}$ concentrations (μg/m$^3$) were determined.
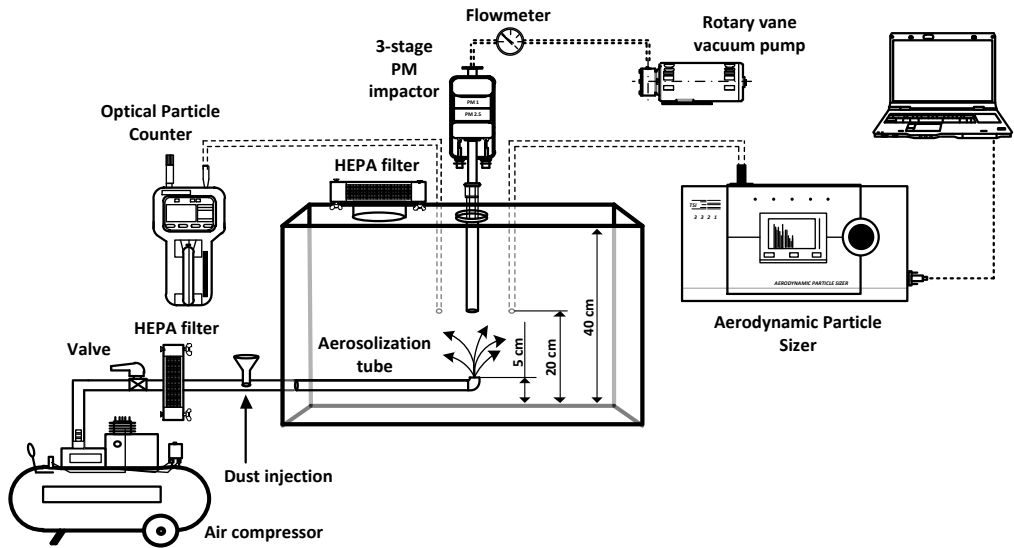
**Figure 2.2.** Experimental system for simulating aerosolization of powdery building materials by fluidization method
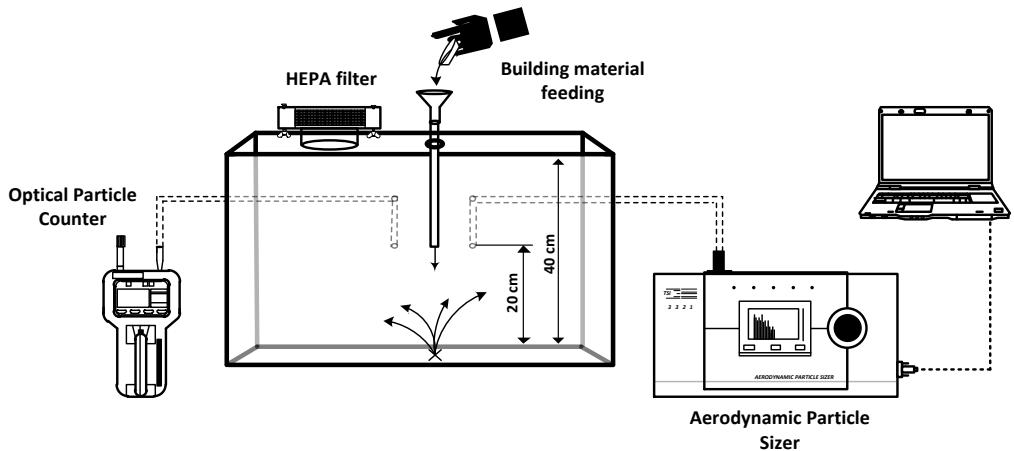


**Figure 2.3.** Experimental system for simulating aerosolization of powdery building materials by gravitation method

## 2.1.2. Aerosol emissions from asbestos and non–asbestos cement roof slates

Samples of three different roof slates were studied. The first sample (further referred to as #1) was prepared from asbestos–cement slate. It was removed from renovated building. The age was approximately 20 years (based on data provided by building owner). The sheet was most likely manufactured in a Soviet Union cement plant. The sheet was well structured, without major cracks or surface corrosion

damages. The second sample (#2) was prepared from a new non–asbestos cement slate, purchased from a building materials store (technical properties: fibrous cement (EN 12467), thickness 6.0±0.5 mm, density >1.40 g/cm$^3$, waterproof, water absorption 30 %). It was produced in the Lithuanian company "Eternit Baltic". The third sample (#3), similarly to the #2 was a non–asbestos slate (technical properties: fibrous cement (EN 12467), thickness 6.5±0.5 mm, density >1.50 g/cm$^3$, waterproof, water absorption ~30 %). It was also purchased from building materials store, and was produced in the Ukraine. Compared to #2, it had more visible and bigger cellulose fibres. All sheets were uncoloured. Before the experiment, each sheet was thoroughly wiped, with the purpose to remove any dust that may interfere with the results. The samples for testing were produced by dividing each slate to pieces of the similar size (approximately 0.07 m$^2$). Each sample was of similar thickness (5 mm). Sheet density was not measured. According to published data, the density of asbestos–cement slate falls in a range of 1.4–2.1 g/cm$^3$ (Gurskis 2008), while the density of non–asbestos slate falls between 1.7–1.9 g/cm$^3$ (EN–12467 2004). The densities of 1.7 g/cm$^3$ of asbestos–cement and non–asbestos slates were used to calculate mass concentrations of PM.

The schematic view of experimental system is presented in **Figure 2.4**. The mechanical operations with sheets were performed in an experimental chamber (553×313×403 mm), which was built from organic (plexi) glass, with the internal volume of 0.061 m$^3$. The interior of the chamber was lined with grounded aluminium foil to prevent electrostatic charge formation causing particle and fibre deposition on the chamber walls. After each operation, aluminium foil lining was removed and replaced with a new one. Air–tight rubber gloves were mounted to the orifices on both sides to provide access for conducting a range of manual sheet–processing operations. A HEPA (class 12) filter was mounted to the orifice on the top of the chamber to ensure clean air supply. The air was withdrawn from the chamber via sampling tubes together with sampled aerosol particles. The movement of air streams in the chamber was modelled using SolidWorks simulation software (version 2010, Dassault Systemes SolidWorks Corp., Concord, MA, USA). This simulation was performed with the aim to ensure equal distribution of air flow streams and low velocities of air streams (<0.1 m/s, representing laminar flow air sampling conditions).

An experiment design involved different treatment operations of asbestos and non–asbestos sheets. Asbestos and non–asbestos roofing slate samples were treated by four different mechanical operations: a) crushing with a hammer; b) rubbing one sheet to another; c) rasping with a file; d) scrubbing with a metal brush. These operations were selected to represent common activities occurring during slate waste removal or cleaning processes. Each sampling run was continued for 5 minutes: a slate sample was processed for 30 seconds period, following by 30 second break, and afterwards repeating 3 more "process–break" cycles.
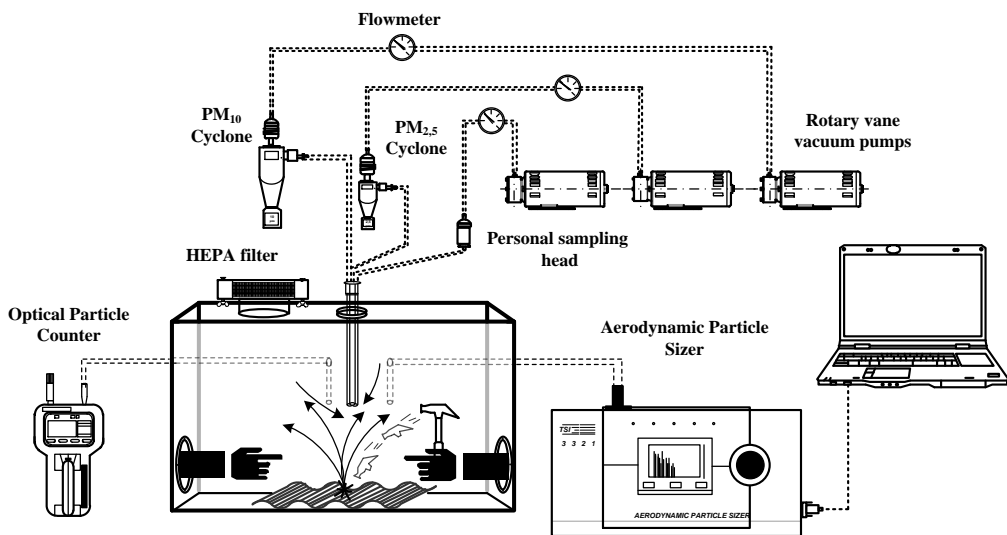
**Figure 2.4.** Experimental system for simulating emissions of PM and fibres from asbestos and non-asbestos cement roof slates

### *Particulate matter sample collection and particle size distribution*

During each run, the generated aerosol from emitted particles and supply air were sampled. Three particulate samples were taken in parallel: a total particulate matter ($PM_{total}$), $PM_{10}$ and $PM_{2.5}$ fractions. Three replicates of each operation were carried out with the aim to estimate the standard error of the experiment. The $PM_{10}$ and $PM_{2.5}$ fractions were separated by cyclones (URG Corporation, Chappel Hill, NC, USA) with appropriate cut–off sizes, at flow rates of 16.7 l/min (1 $m^3$/h). The $PM_{total}$ fraction was collected by diverting the air flow (16.7 l/min) to a 25 mm polypropylene cassette loaded with pre–weighed filter on a steel support pad. Three separate rotary vane vacuum pumps equipped with flow meters were used to maintain the desired sampling flow rates. Aerosolized dust and fibres were collected on mixed cellulose ester (MCE) filters (SKC Inc., diameter 25 mm, pore size 0.8 μm). Filters were conditioned for 24 hours at 20±1 °C before and after sampling. Sample mass was determined by microbalance (model MX5, Radwag, Poland). In total 108 particulate filter samples were taken and analyzed.

In addition to filter–based sampling, several direct–reading instruments were employed. APS and OPC were used in the experiment as well.

The estimation of emission factors of PM and fibres were based on the measured concentration data. Based on samples collected with aerodynamic separators, mass of particles aerosolized from square centimetre of roof slates (sheet fracture line or impact surface) per second ($mg/cm^2/s$) was calculated. The data obtained from APS (total particle number concentration, $PNC_{tAPS}$) and OPC

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($PNC_{tOPC}$) was expressed as number of particles of square centimetre per one second ($\#/cm^2/s$).

With the aim to adequately compare particle emission factors between operations, the surface area of particle emission was estimated for each operation. In case of crushing operation, a fracture line length was measured. For rubbing, rasping and scrubbing operations, the contact area was calculated between the main sheet and another sheet, rasp or metal brush, respectively. Surface area of fracture line of the sheet after crushing operation was determined by the following procedure: initially all edges of the sheet were marked; after the operation, sheet fragments were collected and the length of fracture line was measured. The impact surface area during rubbing, rasping and scrubbing operation was determined identically, i.e. estimating the contact area between the sheets (varying area), sheet and rasp (34 $cm^2$), or sheet and brush (37.5 $cm^2$).

*Mineral fibre counting*

A sample for enumeration of fibres concentration in the aerosol was collected by drawing a known volume of air through a membrane filter by a sampling pump. The aerosolized fibres were collected by special filter cartridge for fibre collection, made from electrically conductive polypropylene (SKC Inc., PA, USA). Filter cartridge was preloaded with MCE membrane filter. The adopted methodology from WHO (1997) was used to determinate airborne asbestos fibre concentration in a chamber (expressed as fibres/cm³). Reflux condenser method was used for preparation of MCE filters for microscopic analysis. Low amount of liquid acetone (250 µl) was stored in three–necked flask; one of the flask holes was installed with reflux condenser. One of the other two holes was plugged, and the third hole was built with two–orifice stopcock for acetone vapour release. The sample (filter mounted on objective slide) was hold 15–25 mm distance from outlet for 3–5 seconds period. Sample was slowly moved across the outlet until it became clear. Triacetin (glycerine triacetate) was used for better fibre contrast.

This method enables measuring number concentration of airborne fibres, defined as objects with a length >5 µm, a width <3 µm and a length–to–width ratio >3:1. Filter was mounted on a microscopic slide and clarified with acetone vapour for light microscopic analysis (Phase Contrast Optical Microscopy, PCOM). We used a positive PCOM with blue filter, an adjustable field iris, ×15 eyepieces, and a 40 phase objective (total magnification of ×600). Random 100 eyepiece grid areas were analyzed. Effective filter area was 385 $mm^2$ and the area of a microscopic field (analysis area) approx 0.038 $mm^2$. Concentration of each individual sample of airborne fibres was expressed as (WHO 1997):

$$C = (A \cdot N) / (a \cdot n \cdot r \cdot t) \qquad (1)$$

here: C – concentration (fibre/$cm^3$); A – effective filter area ($mm^2$); N – total number of fibres counted; a – graticule counting area ($mm^2$); n –number of graticule areas

examined; r – flow rate of air through the filter (ml/min); t – single sample duration (min).

## 2.1.3. Characterization of mineral fibre emissions during building construction/refurbishment and operation phase

*Pilot scale study simulation of fibre emission (Construction/refurbishment phase)*

Experimental setup (pilot scale) (height – 0.7 m; length – 0.3 m; width – 0.3 m) representing the structure of an insulated drywall ceiling was designed for determination of MMVFs penetration into the indoor environment (**Figure 2.5**). Setup was made of galvanized sheet steel, which inner side was sprayed with anti– static solution to eliminate electrostatic forces. Gypsum cardboard plate was attached to standard metal frame. Researched mineral wool was laid down on the top of the frame. Three types of mineral wool were researched, including two types of rock wool (MW1, MW2) and a glass wool (MW3). Air supply nozzle with a valve was installed on the wall of the setup at the level of the mineral wool to model possible situation of an air flow through the layer of insulation. Two nozzles were mounted above and under the gypsum board for pressure difference measurements. Top of the experimental setup was covered with a tight metal cap.
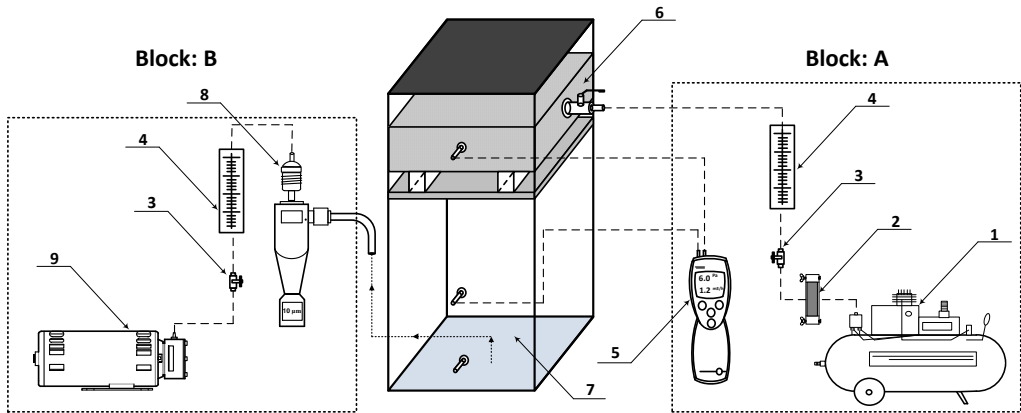


**Figure 2.5.** Pilot scale experimental system designed for determination of MMVFs penetration into the indoor environment
Block A (fibre release): 1 – air compressor; 2 – HEPA filter; 3 – air flow control valve; 4 – rotameter; 5 - pressure meter (testo 512, Testo Limited, Germany); 6 – layer of mineral wool;
Block B: (fibre collection): 7 – fibre collection plate (aluminium foil); 8 – cyclone with special filter cartridge; 9 – rotary vane vacuum pump.

Artificial orifices were created in gypsum board to simulate micro–cracks, providing area for the fibre penetration. One and two millimetre cracks were simulated, with overall area of 10.2 and 40.8 $mm^2$, respectively. In addition, the opening created by a set–in halogen bulb fixture (opening of the width of 3.6 mm, area – 342.5 $mm^2$) was analysed (**Figure 2.6**). The penetration of MMVFs was

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modelled under two conditions – having an air flow through the layer of insulation actively caring fibres through micro–cracks, at a pressure of 6 and 10 Pa. In another case, static conditions with no air flow were researched. Three repeats of each test were performed for quality assurance purposes.
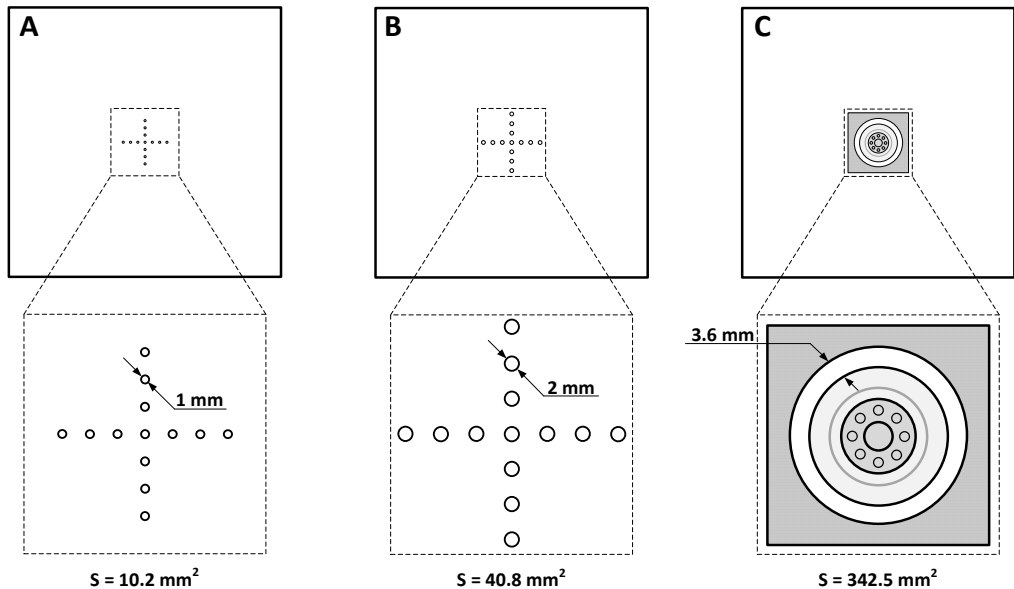


**Figure 2.6.** Three cases of the creation of crack/openings in gypsum cardboard panels (S – total area of cracks, $mm^2$)

The fibres which were released and penetrated through the micro–cracks or openings in the ceiling structure have settled to the floor of the chamber on a sheet of an aluminium foil. After the experiments, the fibres were collected by vacuuming the surface of the foil. The air flow was first directed to the PM10 cyclone (URG Corp., USA) to remove particles, larger than 10 μm in aerodynamic size. The fibres were then collected in a polypropylene cassette loaded with mixed cellulose ester membrane filter. The filters were processed and the fibres were counted according to the methodology from WHO (1997).

### *Temporal variation of fibres in a residential house (Operation phase)*

A recently built low–energy residential house was chosen for the second stage of the study. In this building, the thermal insulation of roofing (35 cm) was accomplished using mineral wool, thus substantial presence of the MMVFs was hypothesized. The ceiling insulation consisted of rock wool (above the moisture barrier) and glass wool (above the gypsum board but below the moisture barrier). Internal wall insulation – rock wool, between gypsum cardboard panels (GCP); floor – reinforced concrete with polyvinyl acetate (PVA) fibres.

The research was aimed at investigating the variation in MMVFs surface densities over time after the completion of building works. The study was carried

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out for 4 months with monthly collection of samples in five different rooms (20 samples collected). Rooms were thoroughly cleaned with a vacuum cleaner before each sampling to represent the removal of fibres via ordinary household activities.

### The presence of MMVFs in multifamily residential buildings (Operation phase)

Determination of MMVFs density in the indoor environment was performed in ten multifamily buildings, which age ranged from 20 to 70 years. These buildings were investigated as future participants in the building refurbishment programme. The buildings were situated in various areas of the city (traffic intensity, population density, etc.) and were distinguished by the structural nature and a year of construction. Five apartments per multifamily building were selected for the research; two Petri dishes were exposed in each apartment. A total number of 100 fibre samples were collected.

### Counting of fibres

MMVFs were collected as a settled dust on a standardized surface (Petri dish), which was exposed indoors for one week, during the second and third stages of the experiment. Afterwards fibre samples were prepared for the analysis under the microscope. Adhesive gel tapes (Dust–lifters, S=14 cm$^2$, BVDA International, the Netherlands) were used to transfer fibres from Petri dish on a microscope slide.

Fibre sample analysis and counting was performed by the PCOM method (microscope Optika B–500 TiPh, Italy, with an integrated camera and software for fibre dimension analysis). This method allows determining not only the surface density of fibres (fibre/cm$^2$), but their structure properties and visually distinguishes mineral fibres from the non–mineral. At least 100 fields were analysed in each sample ($S_{total} = >1$ cm$^2$). This represents 7–8 % of the total area of the gel strip.

MMVF surface density was calculated according the following equation (Valarino et al. 2003):

$$c = N / (S \cdot n) \qquad (2)$$

here: c – MMVFs surface density (fibre/cm$^2$); N – total number of MMVFs counted in one sample; S – area of one analyzed field, cm$^2$; n – number of field areas examined.

## 2.2. Particulate matter in multifamily apartment buildings

### 2.2.1. Particulate matter concentrations in multifamily apartment buildings before refurbishment

*Measurement design*

Ten multifamily buildings (five apartments per building) were included in the measurement campaign during two heating seasons of years 2011 and 2012 (Dec 2011– Apr 2012 and Oct – Dec 2012). Selected multifamily apartment buildings were located in the Kaunas city area. It is the second–largest city in Lithuania (pop. 304 000; total area 158 km$^2$).

The winter time was chosen because of presumably lower impact of outdoor air entering through open windows, more stable thermal conditions indoors (Hanninen et al. 2011), and longer time spent at home by residents. Measurements were performed during working days (Monday–Friday). All buildings were located in the urban areas, affected by traffic pollution from streets. The apartments were selected with the aim to represent different conditions within the building, such as facing south and north directions; upper and lower floor; situated in the middle and corners of the building. Detailed characteristics of the ten case–study buildings are presented in **Table 2.1**.

The number in the parentheses (B#) represents building number and (A#) represents the number of apartment. Corresponding building and apartment coding was selected with the aim to facilitate identification and statistical description of individual apartments and buildings within each over.

**Table 2.1.** General characteristics of ten case study buildings

| Building No. | Year of construction | Type of ventilation | Type of construction | No. of floors | No. of apartments | Total floor area, $m^2$ | Average floor area per apart., $m^2$ |
|---|---|---|---|---|---|---|---|
| **B1** | 1965 | Natural* | Panel Ferro concrete | 5 | 100 | 5270 | 49.5 |
| **B2** | 1957 | Natural* | Brick | 3 | 25 | 2384 | 66.7 |
| **B3** | 1979 | Natural* | Brick | 13 | 60 | 4946 | 61.8 |
| **B4** | 1975 | Natural* | Brick | 12 | 48 | 3726 | 62.1 |
| **B5** | 1992 | Natural* | Brick | 5 | 60 | 3331 | 47.2 |
| **B6** | 1982 | Natural* | Brick | 5 | 51 | 3277 | 54.6 |
| **B7** | 1992 | Natural* | Brick | 5 | 30 | 2056 | 58.3 |
| **B8** | 1981 | Natural* | Brick | 12 | 60 | 3445 | 51.7 |
| **B9** | 1960 | Natural* | Monolithic Ferro concrete | 3 | 30 | 2531 | 59.3 |
| **B10** | 1958 | Natural* | Monolithic Ferro concrete | 3 | 18 | 1344 | 57.5 |

* Most of the apartments had a mechanical exhaust hood above the kitchen stove.

*Measurement methods*

PM concentration and size distribution measurements were performed using OPCs. Direct–reading instruments allowed the estimation of highly time–resolved fluctuations (1 minute averaging) of pollutant concentrations, corresponding to daily activities of inhabitants or other pollution sources. Sampling duration in each apartment was at least 24 hours (full day measurement). PM mass concentrations were calculated based on particle density of 1.5 $g/cm^3$. The density of ambient particles mostly falls within 1.2 to 1.8 $g/cm^3$ (Morawska et al. 1999; Pitz et al. 2003). Particle density is an important parameter affecting the calculation of particle mass from number measurements with OPCs (Tittarelli et al. 2008). In this perspective, the absolute mass concentration values presented in this research study may be biased compared to gravimetric measurements. The OPCs are also known for varying responses to different density, refractive index and shape of particle (Cheng 2008). They are also not capable of detecting nano–sized particles (<0.3 µm). On the other hand, they are compact and lightweight devices providing highly time–resolved particle size distributions and are extensively utilized in indoor and ambient air quality measurement studies (Fittschen et al. 2013; Gorner et al. 2012; Lonati et al. 2011; Shehabi et al. 2008; Weber et al. 2012). Cross–section of cooler box used as an enclose compartment for the PM measurement is illustrated in the **Annex 1**.

In each apartment, measurements were conducted indoors and outdoors. One OPC was set indoors, mostly in the living room, positioned in the area with no primary activities. The indoor sampling height was chosen according to the human breathing zone as seated, i.e. 1.2–1.5 m above the ground. Another OPC was set outdoors in the apartment's balcony or hung outside of the window. The OPCs were enclosed in insulated boxes (specially adapted cooler boxes) with the aim to protect them from environmental stress as well as to protect the inhabitants from the pump noise. Photos from *in situ* measurements are presented in the **Annex 2**.

In parallel to the PM measurements, concentrations of $CO_2$, CO, ambient temperature, relative humidity, and effectiveness of natural ventilation were determined.

Short-term real-time measurements of indoor $CO_2$ and CO concentrations were conducted using IAQ monitors (HD21AB, Delta OHM, Italy). In each apartment, measurements were performed for 24-hour period. Measurement procedure of evaluation of $CO_2$ and CO concentrations was identical to PM concentration assessment, with the exception of the outdoor sampling. Instruments measured $CO_2$ concentrations from 0 ppm to 5000 ppm with an accuracy of ±50 ppm or 3 %.

Two months continuous monitoring of temperature (T) and relative humidity (RH) was initially planned, which was in some cases extended to one year in order to study seasonal variations. Data was recorded with one hour resolution using data loggers (DT-172, Shenzhen Everbest Machinery Industry Co., Ltd, China). These loggers measure temperature from -40 to 70 °C with an accuracy of ±1 °C and RH from 3 % to 100 % with an accuracy of ±3 %. Two loggers per apartment were

placed, one for the coldest spot (i.e. spot with minimum inner surface temperature detected by IR-thermometer, usually by the balcony door) and the other for warm area (e.g., middle of the living room with the height of 1.2-1.5 m above the ground, i.e. human breathing zone as seated). All units used in the study were new and recently calibrated by the manufacturer. Outdoor T (To) and RH (RHo) data were obtained from local monitoring stations during the corresponding period.

The effectiveness of natural ventilation was recorded by an anemometer (417, Testo AG, Germany) by on–spot measuring of air flow velocity through the air vents (bathroom/toilet and kitchen). Natural ventilation was measured twice in each apartment: at the beginning of measurements during the first day visit and after 24 hours period. The vents were usually sized as $0.14 \times 0.18$ m, sometimes having a mechanical blower installed, which operates on a short–term basis. Vents in the kitchen were often connected to the stove exhaust hood. The ventilation rates were calculated as an air change rate (ACH, $h^{-1}$) based on the air flow velocity through the vents and the volume of the apartment. Spearman rank coefficient of correlation (rS) was calculated for the assessment of the relationship between ventilation rate and PM indicators.

Questionnaires and diaries were provided for occupants in addition to active sampling. Questionnaires were constructed to collect information related to apartment information, indoor environmental quality and health. Questions were asked in order to be able to analyse the group level data gathered. The results of the survey were handled with absolute confidentiality so that no information given by individual respondents could be identified. The occupants were asked to fill diaries for the assessment of daily indoor and outdoor activity patterns as well. One occupant per apartment was asked to fill in the questionnaire and diary.

Data on background building information, including energy certificate (energy consumption information), refurbishment plans, architectural and structural designs, and documents related to previous investigations of building structures and their condition were collected.

### Data analysis/Quality assurance

In this study, we have utilized a set of newly obtained factory calibrated OPCs. The six OPCs used in the study were tested side by side under the same conditions after each week of sampling to assess relative bias and precision, followed by purge and sampling flow rate tests. Particle counts within each size category varied by less than 15 % among the OPCs, therefore no correction factors were applied to the analysis of data. Valid measurement results (at least 1320 data points without error flag) were obtained in all apartments, while outdoors 44 cases of valid measurements were collected out of 50. Common reasons for missing data were power failures, malfunctioning of the OPC due to low temperature, or accidental termination of the measurements by the inhabitants.

Data processing and management was carried out using Excel 2007 (Microsoft Inc., USA) and OriginPro 9.0 (Version 9.0, OriginLab Corporation, USA). The

hypotheses on the distribution of the data were tested using the Shapiro–Wilk test (Shapiro and Wilk 1965). In most cases (90 %) the hypothesis on normal distribution was rejected, thus non–parametric tests were further utilized for the data analysis. A one–sample Wilcoxon signed rank test was performed to test if I/O ratios are statistically significantly different from the unity (Wilcoxon 1945). The Mann–Whitney U test was utilized for the determination of significance of difference between nighttime and daytime concentrations (Mann and Whitney 1947).

PM concentration decay rates have shown interaction between ventilation, gravitational settling and penetration from outdoor air. Decay rates were investigated similarly to methods employed by He et al. (2005). Concentration decay rates were determined by the exponential regression fitting of the measured $PM_{2.5}$ concentration decay curves. The selected periods for the determination of the decay had a clear decrease in concentration lasting for a minimum of two hours. The goodness–of–fit of the exponential regression curve was at least 90 % ($R^2 > 0.9$).


## 2.2.2. Particulate matter concentrations before and after refurbishment in multifamily buildings

Measurements of PM concentrations were performed in a total number of seven multifamily buildings (30 apartments, 3–5 apartments per building) before and after refurbishment during three heating seasons of years 2012 and 2014 (Jan – Apr 2012, Oct 2012 – Apr 2013, and Jan – Apr 2014). Building selection principles, as well as PM concentration and size distribution measurements were performed using methods described in the section 2.2.1.

To ensure identical measurement conditions the location of PM measurement devices were positioned in the same places of the apartment before and after the refurbishment. PM measurements were performed after the minimum period of a half year after the full completion of the refurbishment works. The following standard refurbishment works were carried out in all studied multifamily apartment buildings: full facade (including the base) thermal insulation, heating and hot water systems reorganization, reorganization of ventilation and recuperation systems, roof thermal insulation (solar collectors were installed on the top of the roof in some cases), replacement of old windows to new windows with lower thermal conductivity in apartments and other communal areas, glazing of balconies or loggias, replacement of outdoor staircase and tambour doors, etc.

One selected multifamily building before and after the completion of refurbishment works is presented in the **Annex 3**.

## 3. RESULTS AND DISCUSSION

### 3.1. Characterization of particle size distributions of powdery building material aerosol generated by fluidization and gravitation

This part of dissertation aimed to determine PSD of powdery building material aerosol and as an ambient air pollutant (building construction/refurbishment phase). The obtained data were applied to calculate size–segregated emission factors. The results may be utilized to assess potential pollution levels and PSD of fine dust ($PM_{10}$ and $PM_{2.5}$) in either ambient or indoor air during various processes of construction or buildings renovation.

The PSD of resuspended PM from powdery building materials was rather similar obtained either by fluidization or gravitation methods, with an exception of wood sawdust and sand, which varied substantially compared to other materials. In a case of fluidization, wood grinding dust produced most particles in a coarse range (mode equal to 3.28 µm), while gypsum particles were dispersed in a finer range (1.98 µm) according to particle number concentration (**Table 3.1**). In a case of gravitation the highest mode was also assessed for wood grinding dust – 3.52 µm and the lowest – for wood sawdust 0.54 µm.

**Table 3.1.** Value of modes based on number (N) and mass (M) concentration of building materials

| Building material | Fluidization | | Gravitation | |
|---|---|---|---|---|
| | Mode, µm | | Mode, µm | |
| | N | M | N | M |
| Cement | 2.29 | 3.28 | 2.64 | 3.52 |
| Chalk | 2.29 | 3.05 | 2.64 | 13.82 |
| Clay (granulated) | 2.64 | 3.52 | 3.05 | 4.07 |
| Gypsum | 1.98 | 2.84 | 2.29 | 3.05 |
| Hydrated lime | 2.29 | 4.37 | 3.05 | 4.7 |
| Masonry grout | 2.46 | 3.52 | 3.28 | 4.37 |
| Quartz sand | 2.64 | 4.37 | 2.64 | 3.79 |
| Sand | 2.29 | 15.96 | 3.28 | 4.7 |
| Structural lime | 2.13 | 4.37 | 2.64 | 5.05 |
| Wood grinding dust | 3.28 | 5.05 | 3.52 | 5.83 |
| Wood sawdust | 2.64 | 19.81 | 0.54 | 19.81 |

With respect to mass concentration (M), the highest modes by a fluidization method were determined 19.81 µm and 15.96 µm for wood sawdust and sand, respectively. The lowest mode was determined for gypsum – 2.84 µm. In a case of gravitation, the highest modes were assessed for wood sawdust 19.81 µm and chalk

13.82 μm, and the lowest one for gypsum – 3.05 μm. It is clearly visible that PSD by mass concentration retains relatively uniform data during both methods, only chalk and sand have revealed significant different modes.

Comparative analysis of the highest aerosol modes generated by fluidization and gravitation methods revealed a general trend that slightly higher modes of number (N) and mass (M) concentrations were observed for aerosols generated by a gravitation method.

Comparison of PSD (N) of several powder building materials (cement, chalk, gypsum, sand and wood sawdust) during fluidization and gravitation are presented in **Figure 3.1**. It has appeared that both techniques produce slightly different particle size distributions based on the number concentration.

During fluidization and gravitation bimodal particle size distributions were observed mostly by OPC, only during gravitation APS has revealed more tendencies towards the clear bimodal size distribution (chalk – 0.63 and 2.64 μm, sand – 0.54 and 2.29 μm, wood sawdust – 0.54 and 3.52 μm). PSD measured by the APS was slightly shifted compared to OPC in all measurements. The differences were caused by different measuring ranges and resolutions of the instruments (OPC starts to measure from 0.3 μm, APS from 0.5 μm), and possibly due to different classification techniques of particles (optical vs. aerodynamic/optical).

In some cases of PSD by OPC (cement and wood sawdust), mostly from fluidization, the modes were visible at >10 μm channel (increase of PM). This could be caused due to the high PM concentration in the chamber approaching an upper range of the instrument operation that brings discrepancies in measurement results, e.g., two 5 μm particles passing throughout the laser beam of an instrument could be identified as a one 10 micrometer particle. Therefore, this method is not always accurate for such experiments when PM concentrations approach an upper range of the instrument. In such cases a PM impactor which classifies particles aerodynamically may be utilized.

In this study the $PM_{10}$ portion (as determined by 3–stage impactor measurements) amounted between 29.5 % and 86.6 % (quartz sand and masonry grout, respectively), the $PM_{2.5}$ portion between 6.6 % and 28.1 % (quartz sand and clay, respectively) of the $PM_{total}$ emissions. The $PM_{1.0}$ portion constituted between 0.3 % and 6.5 % (chalk and clay, respectively) of the $PM_{total}$ emission (**Table 3.2**).

$PM_{10}$ fraction mostly composed a significant part of all particles of aerosol (~70–75 %) with an exception of quartz sand and wood dust which revealed the lowest $PM_{10}$, $PM_{2.5}$ and $PM_{1.0}$ ratios compared to $PM_{total}$. At the same time, $PM_{2.5}$ fraction represents a relatively small proportion of total aerosolized PM (~15–20 %). On the other hand, this finding shows that although the tested methods generate a substantial amount of fine aerosol fraction, but if inhaled, it may travel to the deepest pathways of a respiratory system and cause associated health effects. It may be concluded that during construction work or structural renovation of buildings substantial quantities of fine PM are emitted into the ambient air and can cause actual health problems.

In comparison to other studies, such as Ehrlich et al. (2007), during this study the majority of analyzed building materials emitted rather similar concentrations of $PM_{10}$ but with different portions of $PM_{10}$ of the TPM emission. The $PM_{10}$ emissions from industrial plants in Germany amounted to more than 90 % and the $PM_{2.5}$ portion between 50 % and 90 % of the $PM_{total}$ emission.

**Table 3.2.** Fractional composition of particulate matter aerosol (by mass concentration, in % of $PM_{total}$)

| Building material | $PM_1$ | $PM_{2.5}$ | $PM_{10}$ |
|---|---|---|---|
| Cement | 1.3 | 13.3 | 76.6 |
| Chalk | 0.3 | 12.1 | 78.7 |
| Clay (granulated) | 6.5 | 28.1 | 51.1 |
| Gypsum | 1.6 | 16.1 | 81.1 |
| Hydrated lime | 0.6 | 11.6 | 79.6 |
| Masonry grout | 0.4 | 13.2 | 86.6 |
| Quartz sand | 0.5 | 6.6 | 29.5 |
| Sand | 2.8 | 22.7 | 49.8 |
| Structural lime | 1.3 | 11.7 | 75.7 |
| Wood grinding dust | 1.4 | 9.9 | 35.7 |
| Wood sawdust | 1.0 | 8.8 | 38.3 |

The methods utilized in the above presented experiments for the particle aerosol generation are laboratory based. They are created to mimic closely the process of a scaled–down industrial pollution event. It may be expected that the industrially produced aerosol will have a similar PSD, if the same type of material is used and no pollution control devices are installed between aerosol generation and sampling sites. Of course, the mass and number of generated particles will depend on the mass flow of material. i.e., the emission factors expressed as g/s will be substantially higher compared to laboratory conditions.
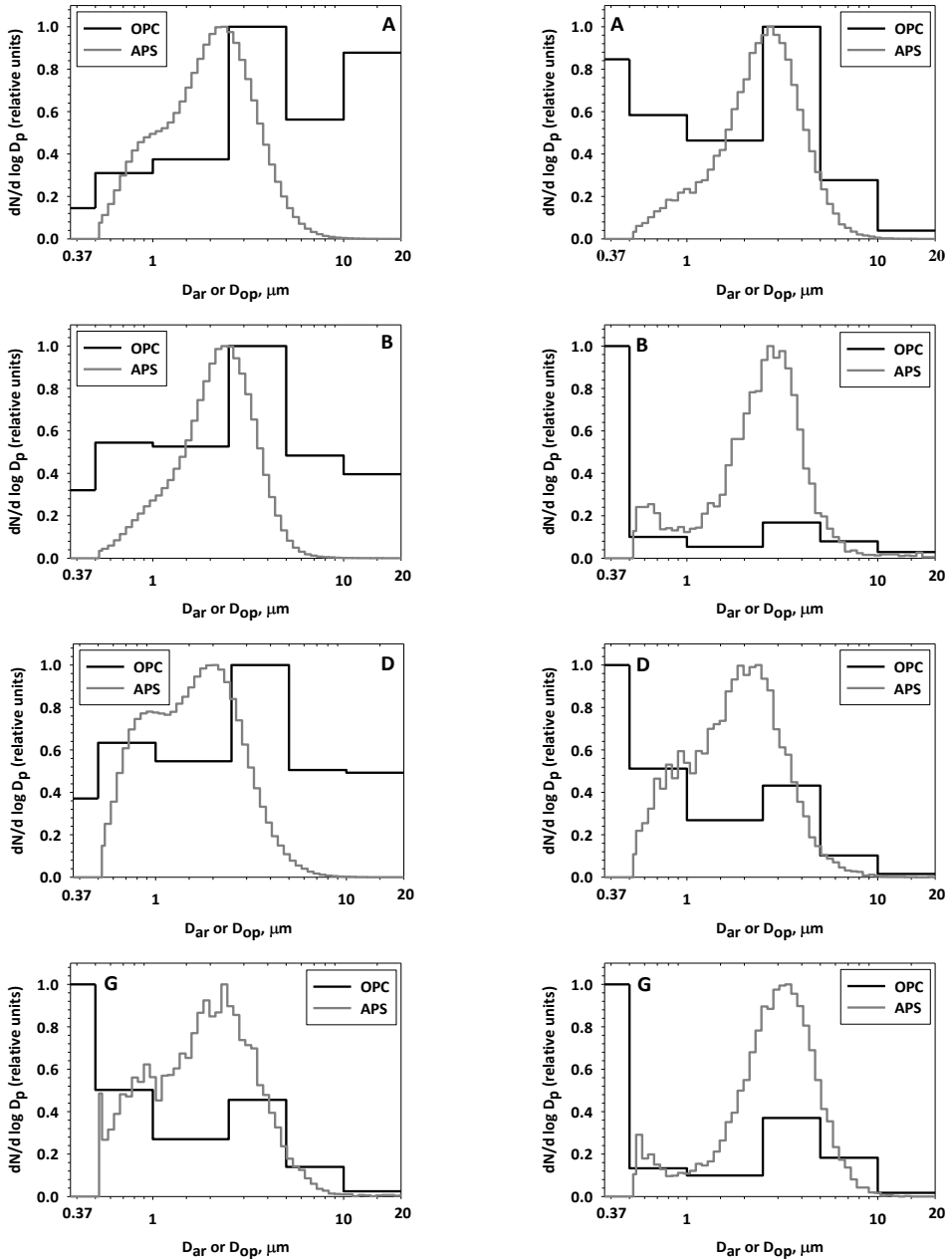
**Figure 3.1.** Particle size distribution (dN) of a) cement, b) chalk, d) gypsum, g) sand, during fluidization and particle size distribution (dN) of a) cement, b) chalk, d) gypsum, g) sand, during gravitation by APS

### 3.2. Comparative characterization of particle emissions from asbestos and non-asbestos cement roof slates

Based on the above described research, this study has focused on two major aspects. We aimed in characterising not only fibres, but total and size–fractionated PM occurring from the processing of cement roofing slates. This has been motivated by the fact that the total aerosol from cement roof slating has not been characterized from particle size distribution perspective, while not only fibres, but cement particles may also be of potential hazard to human health. Long–term exposure to airborne crystalline silica can cause a lung disease – silicosis. A frequent cause of death in people with silicosis is pulmonary tuberculosis (silico–tuberculosis) (Silicosis 2000).

Moreover, we aimed in characterizing particle emissions not only from asbestos–cement sheets, but also from new non–asbestos cement sheets, with respect to total and size–fractionated particulate matter as well as cellulose fibres emissions. The research provides new insights in the assessment of potential impacts of roof slates to urban air pollution and human exposure to pollutants.

Chapter 3.2 further is divided in two sections were particle emission factors (section 3.2.1) and particle size distribution of generated PM (section 3.2.2) during various mechanical operations are observed.

### 3.2.1. Characterization of particle emission during various mechanical operations

One of the aims of this study was to assess total PM emission from cement sheets. The release of particles was estimated and compared for different types of sheets and operations.

The experimental data on particle release is summarized in **Table 3.3**. A substantial variation in the mass concentration of generated aerosol was noticed, both within each PM fraction and between different operations. The highest aerosolization of particles was observed during different operations: sample #1 – scrubbing, #2 – crushing, #3 – rubbing; the lowest aerosolization of particles occurred during different operations: sample #1 – rasping, #2 – rubbing, #3 – scrubbing. Crushing appeared as the most fibres emitting operation, producing highest concentrations of fibres from all tested sheets (both asbestos and non-asbestos cement).

Distribution of PM number concentration in time for all mechanical operations of different types of sheets is presented in **Annex 4**.

Particle release expressed by number concentration was assessed by two types of instruments – APS and OPC. As it was expected, the OPC yielded higher values of concentration due to lower detection limit with respect to particle size. Similarly to mass concentration, the highest emission factors were assessed during crushing operation (**Table 3.3**); the minimum emissions were during rubbing of sample #1.

With respect to mass concentration, the $PM_{total}$ fraction, naturally, contained the greatest amount of PM in all runs. The ratios of PM concentrations between the

fractions ($PM_{10}/PM_{total}$) from #1 sheet for crushing, rubbing, rasping and scrubbing were 0.88, 0.84, 0.64 and 0.92 respectively. In dust from #2 sheets the ratios were 0.88, 0.84, 0.98 and 0.88; in #3 – 0.76, 0.85, 0.92 and 0.71 respectively for all operations. This shows that $PM_{10}$ fraction composed a significant part of all particles of aerosol (~70–98 %). The ratios $PM_{2.5}/PM_{total}$ from #1 sheet for crushing, rubbing, rasping and scrubbing were 0.11, 0.18, 0.22 and 0.17 respectively. In dust from #2 sheet ratio were 0.14, 0.31, 0.31 and 0.25; in #3 – 0.14, 0.40, 0.16 and 0.15, respectively for all operations.

Hence, $PM_{2.5}$ fraction composed a small part of total aerosolized PM (~20 %). On the other hand, this finding shows that although the tested operations are mechanical, a substantial amount of fine aerosol fraction is generated, and, if inhaled, may travel to the deepest pathways of respiratory system and cause associated health effects.

**Table 3.3.** Emission factors of $PNC_{tOPC}$, $PNC_{tAPS}$ (#/cm$^2$/s) and PM fractions expressed as mg/cm$^2$/s

| Operation title | Sheet type | $PNC_{tOPC}$, #/cm$^2$/s | $PNC_{tAPS}$, #/cm$^2$/s | $PM_{total}$, mg/cm$^2$/s | $PM_{10}$, mg/cm$^2$/s | $PM_{2.5}$, mg/cm$^2$/s |
|---|---|---|---|---|---|---|
| Crushing | #1 | 771.1±105.3 | 390.0±62.1 | 2.63E-04± 1.15E-04 | 2.27E-04± 1.66E-04 | 2.88E-05± 9.79E-06 |
| | #2 | 1672.0±168.4 | 1209.1±103.5 | 7.51E-04± 1.41E-04 | 6.64E-04± 1.53E-04 | 1.04E-04± 2.08E-05 |
| | #3 | 1990.6±201.2 | 1741.9±140.9 | 9.93E-04± 6.38E-05 | 7.53E-04± 1.65E-04 | 1.36E-04± 4.61E-05 |
| Rubbing | #1 | 0.13±0.03 | 0.04±0.008 | 5.32E-08± 8.17E-09 | 4.33E-08± 2.02E-08 | 9.34E-09± 6.13E-10 |
| | #2 | 0.72±0.11 | 0.53±0.06 | 9.01E-08± 1.51E-08 | 7.60E-08± 9.90E-09 | 2.76E-08± 3.13E-09 |
| | #3 | 3.34±0.52 | 2.91±0.36 | 5.74E-07± 5.60E-08 | 4.90E-07± 7.87E-08 | 2.32E-07± 7.16E-08 |
| Rasping | #1 | 1.38±0.31 | 1.14±0.18 | 8.78E-07± 1.75E-07 | 6.34E-07± 4.07E-07 | 3.53E-07± 7.75E-08 |
| | #2 | 8.97±1.12 | 6.11±0.53 | 3.68E-06± 6.20E-07 | 3.44E-06± 8.19E-07 | 1.07E-06± 1.22E-07 |
| | #3 | 11.81±1.96 | 6.57±0.42 | 4.93E-06± 6.02E-07 | 4.54E-06± 9.03E-07 | 8.12E-07± 1.91E-07 |
| Scrubbing | #1 | 9.31±1.02 | 4.80±0.32 | 4.32E-06± 1.24E-06 | 3.86E-06± 1.39E-06 | 8.78E-07± 3.51E-07 |
| | #2 | 11.49±1.16 | 7.35±0.61 | 2.18E-06± 3.93E-07 | 1.92E-06± 1.43E-07 | 5.36E-07± 1.07E-07 |
| | #3 | 16.39±1.86 | 14.41±0.97 | 4.94E-06± 8.33E-07 | 3.51E-06± 8.93E-07 | 7.62E-07± 2.26E-07 |

The results indicate that non–asbestos cement sheets produced higher amount of aerosol particles during all operations. During crushing operation, the emission of particles was 2–3 times higher from non–asbestos cement; for rubbing and rasping – even higher difference. Scrubbing of sample #2 (Lithuanian–made non asbestos sheet) behaved somewhat differently with respect to $PM_{total}$ emissions, but overall, asbestos cement emitted lower amount of particles during mechanical treatment. This may be explained by differences in the structural matrix of cement sheets.

The obtained specific emission factors may be further recalculated to estimate the release of particles from the entire slate. The emissions of particles during crushing operation may be assessed by measuring/estimating an approximate fracture line length and taking one second as a process time (for a single sheet). For other operations, the emissions may be calculated according to the area processed and the duration of processing.

## 3.2.2. Characterization of particle size distribution of generated aerosol during various operations

Utilization of particle size–selective instrumentation allowed the characterization of PSD of the generated aerosol. **Figure 3.2** shows particle size distribution calculated from the number concentration (dN/dlogDp) of the aerosol during the crushing operation of the asbestos–cement sample (#1), plotted using the data from APS and the OPC. Both instruments have revealed bimodal particle size distribution. PSD measured by the OPC was slightly shifted compared to APS. The first mode was measured at 0.3–0.5 μm by OPC, and at 0.6–0.8 μm by APS; the second mode was located at 2.5–5 μm and 1.5–2.5 μm, respectively. The differences were caused by different measuring ranges and resolutions of the instruments, and possibly due to different classification techniques of particles (optical vs. aerodynamic/optical). From the perspective of occupational health, a fine particle mode is important, because it shows significant presence of respirable particles.

Moreover, a substantial amount of asbestos fibres may reside in this mode, due to their small aerodynamic diameter. Another mode occurred in the range of 2–5 μm (thoracic fraction), which is also of a very high hazard with respect to penetration to human respiratory tract (Laden et al. 2006).
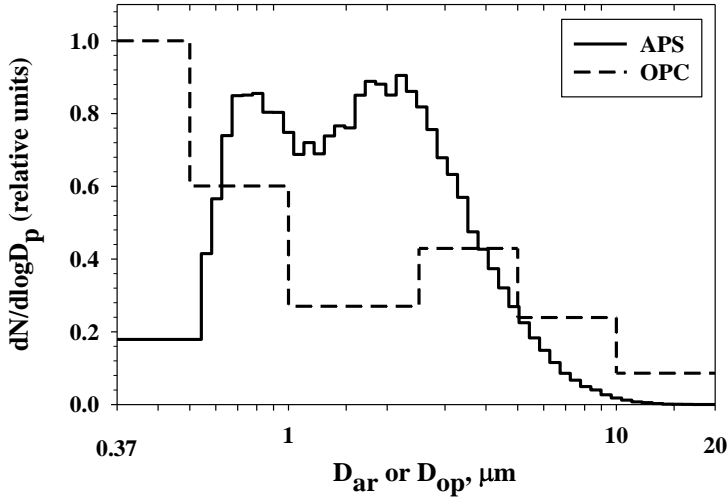
**Figure 3.2.** Particle size distribution (number) of #1 sheet during crushing operation by APS and OPC (particle size relative concentration after normalizing for particle diameter)

PSD based on mass concentration (dM/dlogDp) of an asbestos cement sample, during crushing operation is presented in **Figure 3.3**. It was determined using the data from APS and aerodynamic separators – cyclones. The particles have appeared to be distributed in a single–mode pattern. The modes agreed well and were situated a coarse range (2.5–10 µm). In general, most of generated aerosol fell in the range of thoracic fraction.
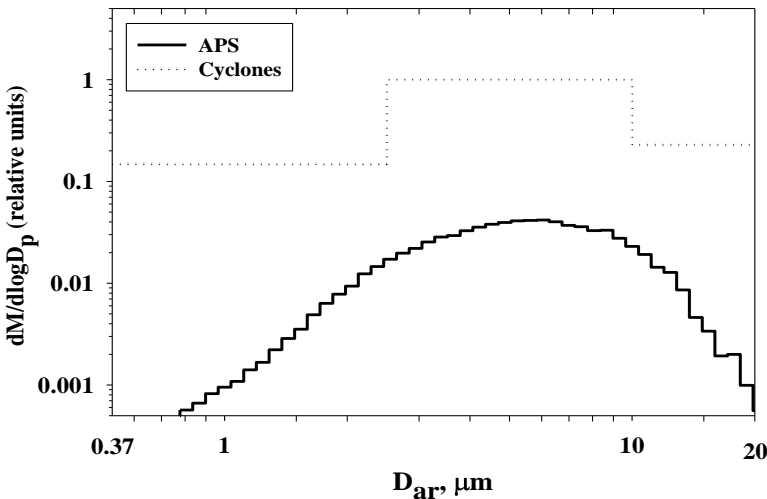


**Figure 3.3.** Particle size distribution (mass) of #1 sheet during crushing operation by APS and filter sampling (particle size relative concentration after normalizing for particle diameter)

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Figures 3.4 and 3.5 compare PSD of the aerosol, generated from all three tested sheets during various operations, as measured with the APS. The quantitative parameters of distributions are presented in Table 3.4.

Similarly to the sample #1 during crushing operation, the non–asbestos sheet samples produced aerosol of bimodal PSD with respect to number concentration, and single mode PSD with respect to mass concentration. For both non–asbestos sheet samples, particles were distributed in a rather similar pattern, while the PSD shapes differed from those of the asbestos cement sample. The shapes of PSDs of sample #1 during rubbing, rasping and scrubbing operations substantially differed from #2 and #3 respective operations. For example, in case of rasping operation of the asbestos sample, the mode value was the lowest and equal to 0.78 μm, while for non-asbestos sheets – 2.13 μm. The maximum particle median diameter (based on number concentration) was assessed during scrubbing of the #3 sample (1.87 μm), the minimum – during rubbing of the sample #3 (1.34 μm). Based on mass concentration, the maximum median diameter occurred during crushing of the #1 sample (5.15 μm), the minimum – during rubbing of the #3 sample (2.97 μm).
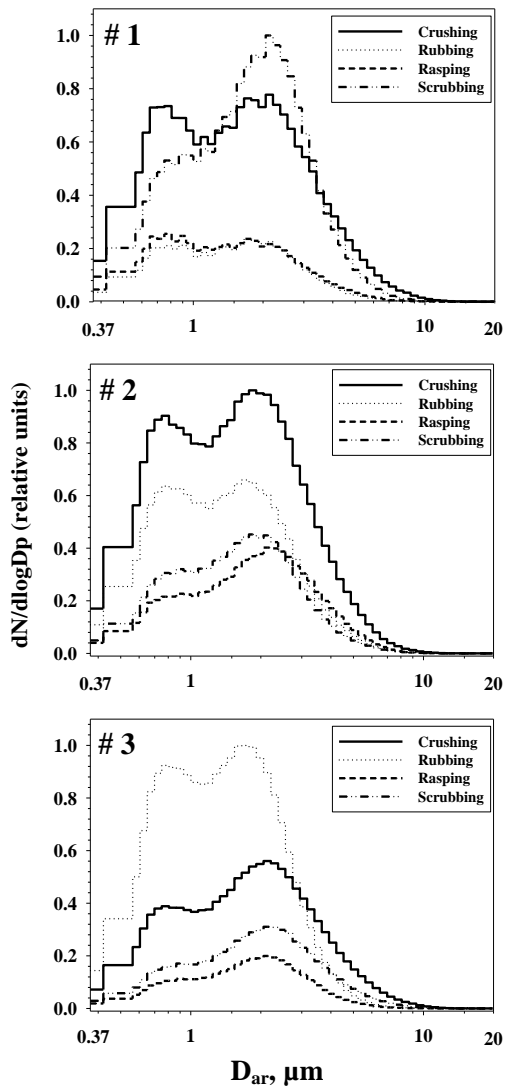
**Figure 3.4.** Particle size distribution by number measured with APS (dN/dlogDp) of different sheets during various treatment operations
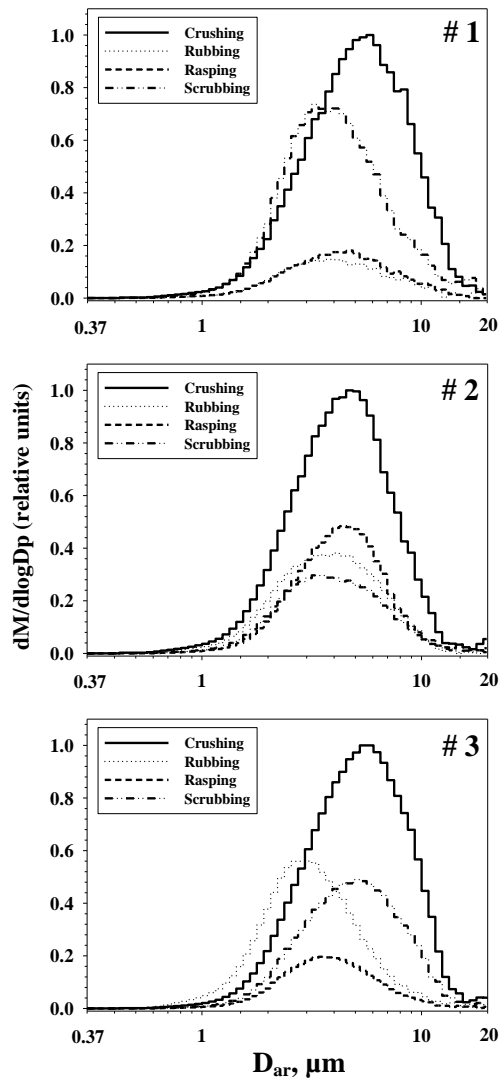
**Figure 3.5.** Particle size distribution by mass calculated from APS data (dM/dlogDp) of different sheets during various treatment operations

**Table 3.4.** Particle size distribution parameters of generated aerosol

| Operation title | Sheet type | Mode, μm | Median, μm | Geo. Mean, μm | Geo. Std. Dev. | $dN/dlogD_p$, #/cm$^2$ | Mode, μm | Median, μm | Geo. Mean, μm | Geo. Std. Dev. | $dM/dlogD_p$, μg/cm$^2$ |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | | Number concentration | | | | | Mass concentration | | | | |
| Crushing | #1 | 2.13 | 1.58 | 1.58 | 1.90 | 0.094 | 5.83 | 5.15 | 4.96 | 1.72 | 1.64E-06 |
| | #2 | 1.84 | 1.48 | 1.48 | 1.81 | 0.308 | 4.70 | 4.07 | 3.92 | 1.66 | 3.94E-06 |
| | #3 | 2.13 | 1.70 | 1.69 | 1.85 | 0.468 | 5.43 | 4.77 | 4.55 | 1.66 | 8.58E-06 |
| Rubbing | #1 | 1.72 | 1.46 | 1.46 | 1.78 | 1.21E-05 | 3.79 | 3.99 | 4.04 | 1.78 | 1.16E-10 |
| | #2 | 1.72 | 1.36 | 1.36 | 1.74 | 1.42E-04 | 4.07 | 3.67 | 3.42 | 1.67 | 1.04E-09 |
| | #3 | 1.72 | 1.34 | 1.34 | 1.68 | 8.34E-04 | 2.84 | 2.97 | 2.94 | 1.64 | 4.82E-09 |
| Rasping | #1 | 0.78 | 1.39 | 1.43 | 1.81 | 3.10E-04 | 4.70 | 4.22 | 4.16 | 1.75 | 2.74E-09 |
| | #2 | 2.13 | 1.84 | 1.74 | 1.81 | 1.84E-03 | 4.37 | 4.22 | 4.05 | 1.62 | 2.77E-08 |
| | #3 | 2.13 | 1.73 | 1.64 | 1.73 | 2.14E-03 | 3.52 | 3.57 | 3.59 | 1.61 | 2.19E-08 |
| Scrubbing | #1 | 2.13 | 1.77 | 1.69 | 1.75 | 1.58E-03 | 3.28 | 3.91 | 4.02 | 1.72 | 1.55E-08 |
| | #2 | 1.84 | 1.58 | 1.52 | 1.73 | 2.23E-03 | 3.28 | 3.50 | 3.55 | 1.66 | 1.90E-08 |
| | #3 | 2.13 | 1.87 | 1.80 | 1.81 | 4.35E-03 | 5.05 | 4.69 | 4.55 | 1.67 | 7.02E-08 |

The presented results show that mechanical operations of both asbestos and non–asbestos roofing slates produce significant amounts of fine aerosol particles (respirable and thoracic fractions), which can penetrate deep into the human respiratory tract and may be potentially hazardous to health. Interestingly, the new generation non–asbestos sheets generate higher aerosol concentrations during mechanical treatment, thus may be of potentially higher hazard with respect to exposure to cement particles, without assessing fibre emissions.

### 3.2.3. Characterization of fibre release

*Aerodynamic classification of asbestos fibres*

The hazardousness of asbestos–cement sheets is most often characterized by the release of asbestos fibres. Usually, total concentration of fibres is assessed. On the other hand, earlier studies have implicated that cement particles may be binding asbestos fibres, thus changing their aerodynamic diameter (Bridle and Stone 2006; Deruyterre et al. 1980). We aimed in classifying fibres emitted during mechanical asbestos treatment (section 2.1.2) by aerodynamic classifiers ($PM_{2.5}$ and $PM_{10}$ cyclones), with the aim to research their potential to penetrate human respiratory tract. The estimation of concentration of asbestos fibres (fibre/cm$^3$), fibre length (µm) and asbestos fibre emission factor (f/cm$^2$/s) were based on measurement and counting results.

Similarly to the total PM release, the highest fibre concentration occurred during crushing asbestos–cement sheets (**Table 3.5**). The lowest emission factor (concentration adjusted for contact area and time) of fibres was calculated during the rubbing operation – almost 1000 times lower than that of crushing. Studies conducted by the National Roofing Contractors Association (NRCA) from 1986 to 1991 indicated that removal of asbestos–containing roof flashings, mastics, coatings and cements yielded low asbestos fibre concentrations (range 0.004–0.027 fibre/cm$^3$; mean 0.024 fibre/cm$^3$) (Mowat et al. 2007). In this study, concentrations of similar order of magnitude were registered: crushing of asbestos–cement sheets formed few times higher concentrations ($PM_{total}$ – 0.051±0.008 fibre/cm$^3$), whereas during rubbing operation, the concentrations fell in the same range ($PM_{total}$ – 0.015±0.004 fibre/cm$^3$).

**Table 3.5.** Asbestos fibre concentration (fibre/cm$^3$), fibre length (μm), and emission factor (EF, f/cm$^2$/s) (±SD) of asbestos–cement sample during various treatment operations

|  |  | **PM$_{2.5}$** | **PM$_{10}$** | **PM$_{total}$** |
|---|---|---|---|---|
| **Crushing** | Fibre conc., fibre/cm$^3$ | 0.059±0.011 | 0.057±0.020 | 0.051±0.008 |
|  | Fibre length, μm | 49.76±8.14 | 52.25±7.56 | 52.34±2.19 |
|  | EF, f/cm$^2$/s | 2.34E-02±4.15E-03 | 2.26E-02±7.77E-03 | 2.01E-02±2.99E-03 |
| **Rubbing** | Fibre conc., fibre/cm$^3$ | 0.025±0.010 | 0.016±0.002 | 0.015±0.004 |
|  | Fibre length, μm | 32.62±3.80 | 41.56±1.17 | 40.10±5.35 |
|  | EF, f/cm$^2$/s | 3.54E-06±1.43E-06 | 2.29E-06±2.62E-07 | 2.06E-06±5.35E-07 |
| **Rasping** | Fibre conc., fibre/cm$^3$ | 0.035±0.008 | 0.031±0.006 | 0.036±0.002 |
|  | Fibre length, μm | 25.28±0.67 | 34.87±0.53 | 37.28±5.92 |
|  | EF, f/cm$^2$/s | 2.05E-05±5.01E-06 | 1.85E-05±3.61E-06 | 2.15E-05±1.36E-06 |
| **Scrubbing** | Fibre conc., fibre/cm$^3$ | 0.025±0.002 | 0.019±0.002 | 0.015±0.001 |
|  | Fibre length, μm | 24.42±5.19 | 32.04±3.22 | 36.32±7.55 |
|  | EF, f/cm$^2$/s | 2.17E-04±1.79E-05 | 1.58E-04±1.58E-05 | 1.31E-04±5.97E-06 |

An interesting phenomenon was observed during classification of asbestos fibres into PM$_{2.5}$, PM$_{10}$ and PM$_{total}$ fractions. It was hypothesized, that PM$_{2.5}$ fraction would contain the smallest amount of fibres, especially if the fibres were bound to cement particles, which in turn would increase their aerodynamic diameter. Bridle and Stone (2006) raised the argument that all chrysotile in asbestos–cement sheet is replaced chemically and structurally, that it would no longer be regarded as pure chrysotile. It was suggested calling these adhered fibres "Casitile" because of calcium (Ca) and silicon (Si) influence. It was noticed a lot of agglomerates on the filters in almost all samples of PM total fraction. In PM$_{2.5}$ and PM$_{10}$ fractions only a small part (~10 %) of agglomerated fibres were detected. Thus, there was no evidence to support claims that all the chrysotile asbestos in asbestos–cement is significantly adhered so that fibres present or released should no longer be considered to be chrysotile. Moreover, for each operation (except rasping) the concentration of asbestos fibres in PM$_{2.5}$ fraction was found to be greater than PM$_{10}$ and PM$_{total}$ fractions. This might have been caused by several experimental issues. Firstly, in PM$_{10}$ and PM$_{total}$ fractions, the particle concentration was substantially higher, thus a portion of asbestos fibres might have been covered with particulate matter and not visible during microscopic analysis. On the other hand, some disaggregation of fibre–cement agglomerates might have occurred in the cyclones,

during inertial separation process. It must be noticed, that the fibre length in $PM_{2.5}$ fraction was the smallest among all three fractions, and only few agglomerates were registered. In any case, the appearance of significant amounts of asbestos fibres in $PM_{2.5}$ fraction confirms high potential hazard of asbestos cement sheets.

Pastuszka (2009) has expressed asbestos fibre emission factor according to the energy, required to perform mechanical process (crushing). The emission rate was defined as a number of fibres (F) emitted from the unit area ($m^2$) of a material due to the impact of unit energy (J). Emission factor was expressed in $F/(m^2{\times}J)$. In order to compare the results with the above mentioned study, we have re–calculated the emission factor for crushing operation, estimating the effect of the force generated by the hammer to the surface area of the slate. The estimated energy of the hammer fall during crushing operation was equal to 9 J. The averaged value of the surface emission rate was equal to $4.9{\times}103{\pm}0.8{\times}10^3$ $F/(m^2{\times}J)$ for fibres longer than 5 µm. This number agrees well with Pastuszka's calculated values of $2.7–6.9{\times}10^3$ $F/(m^2{\times}J)$ (fibres longer than 5 µm) for sheets with either good or damaged surface. This comparison also serves as a good validation of the obtained results.

### *Fibre release from asbestos vs. non asbestos sheets*

One of the main tasks was to compare the release of fibres from asbestos cement and non–asbestos (cellulose) cement sheets. Currently, there is little data available on long–term inhalation of cellulose fibres on human health (Warheit et al. 2001). It was shown that cellulose fibres can have adverse effects on the airways (Cullen et al. 2002; Cullen et al. 2000). Although the potential hazard of cellulose fibres to human respiratory system is considered as substantially lower than that of asbestos. It is feasible to research the emissions of these fibres from non–asbestos cement sheets, in order to determine the emission factors, which may further be utilized for epidemiological studies.

The results revealed that the emission of cellulose fibres was indeed lower, compared to asbestos fibres. During crushing operation, the emission of fibres was 3.2 times higher compared to sample #2, and 7.4 times higher from sample #3 perspective. For rubbing the sample #1 concentration ratios were 3.5 and 1.5, rasping – 4.3 and 13, scrubbing – 3.5 and 1.8 higher in comparison with sample #2 and #3, respectively. Also, an average cellulose fibre was shorter compared to asbestos fibre (**Table 3.6**). The average fibre length was approximately 1.4 and 1.6 times lower for samples #2 and #3, respectively. It may be concluded, that during mechanical treatment operations of non–asbestos cement sheets, substantial quantities of cellulose fibres are emitted into ambient air. The obtained fibre concentration in chamber's air was relatively low. On the other hand, the human respiratory flow rate (~30 l/min) is higher than the flow rate which was used for sampling (2.2 l/min), thus the fibres may have been aspirated with lower efficiency, compared to human inhalation. In case of long term worker exposure, the dose may approach that of tested by Cullen et al. (2002). Based on these findings, it is difficult to draw firm conclusions on the potential hazard of non–asbestos sheets to worker's

health. The obtained data should be reviewed with the appearance of new data about the effect of cellulose fibres on human health.

**Table 3.6.** Asbestos and cellulose fibres release during various mechanical operations

|  |  | # 1 (asbestos) | # 2 (cellulose) | # 3 (cellulose) |
|---|---|---|---|---|
| **Crushing** | Fibre conc., fibre/cm$^3$ | 0.036±5.7E-03 | 0.011±1.3E-03 | 0.005±8.5E-04 |
|  | Fibre length, μm | 30.34±3.59 | 30.00±2.00 | 18.75±1.36 |
| **Rubbing** | Fibre conc., fibre/cm$^3$ | 0.010±2.1E-03 | 0.003±6.9E-04 | 0.007±6.9E-04 |
|  | Fibre length, μm | 34.37±4.12 | 25.05±3.58 | 23.20±4.10 |
| **Rasping** | Fibre conc., fibre/cm$^3$ | 0.011±1.9E-03 | 0.002±1.2E-03 | 0.001±7.0E-04 |
|  | Fibre length, μm | 17.78±1.33 | 17.50±1.85 | 12.50±2.12 |
| **Scrubbing** | Fibre conc., fibre/cm$^3$ | 0.012±2.1E-03 | 0.003±3.5E-04 | 0.007±6.1E-04 |
|  | Fibre length, μm | 32.22±5.06 | 15.00±2.36 | 18.00±3.14 |

## 3.3. Characterization of mineral fibre emissions during building construction/refurbishment and operation phase

The specific aim of this chapter was to assess MMVFs emissions and fibre pathways into indoor environment during building construction/refurbishment and operation phases.

Characterization of MMVFs emissions was divided into three stages: 1) assessment of fibre emissions from various insulation materials under laboratory conditions (construction/refurbishment phase); 2) identification of mineral fibre surface density after the construction and interior installation of the case–study building (operation phase); and 3) determination of mineral fibre surface density in the indoor environment of selected multifamily buildings (operation phase).

### 3.3.1. Assessment of fibre emissions from various insulation materials in pilot scale experimental system

The air flow through the layer of insulation resulted in the increased emissions of MMVFs (**Table 3.7**). MMVF emissions varied in the range of 7–44 fibre/cm$^2$/h under air flow conditions and were dependent on the type of the mineral wool. MMVF emissions under static conditions ranged from 2.5–5 fibre/cm$^2$/h and, in some cases, the MMVFs were not detected at all. MMVF emissions varied depending on the micro–crack size as well. MMVF emissions were 7–23, 11–30,

and 13–45 fibre/cm$^2$/h when micro–cracks were 1.0, 2.0, and 3.6 mm in diameter, respectively. An under–pressure of 10 Pa revealed higher MMVF emission comparing to 6 Pa under–pressure. Such tendency was observed with all types of mineral wool. Laboratory study has shown that fibre penetration were comparatively high even at the minimum area of micro–cracks.

The obtained results indicate that during almost all researched scenarios, there is a potential to reach the threshold value of 10 fibres/cm$^2$ (value designated for occasionally cleaned surfaces) over prolonged periods of time. The exceedance of the threshold value calls for corrective actions, such as increased cleaning and/or considering improved cleaning methods; elimination of source followed by thorough cleaning (Schneider 2000). If possible, the surface area of crack should be minimized, and the overpressure conditions should be avoided by the installation of the wind barriers in roof insulation.

**Table 3.7.** Emissions of MMVFs from different mineral wools in experimental setup

| Mineral wool type | Total area of micro cracks, mm$^2$ | Pressure difference, Pa | Supplied air flow, l/min | Mean MMVFs emission, fibre/cm$^2$/h |
|---|---|---|---|---|
| MW1 (Rock wool) | 10.2 | - | - | 2.5 |
| | | 6 | 8 | 19.0±12.3 |
| | | 10 | 14 | 23.0±7.1 |
| | 40.8 | - | - | 2.5 |
| | | 6 | 11 | 21.0±10.6 |
| | | 10 | 16 | 30.0±14.1 |
| | 342.5 | - | - | 5.0 |
| | | 0.3 | 8 | 41.0±3.5 |
| | | 0.8 | 15 | 45.0±7.0 |
| MW2 (Rock wool) | 10.2 | - | - | 0.0 |
| | | 6 | 8 | 12.0±8.8 |
| | | 10 | 11 | 17.0±1.8 |
| | 40.8 | - | - | 2.5 |
| | | 6 | 9 | 17.0±5.3 |
| | | 10 | 15 | 20.0±1.8 |
| | 342.5 | - | - | 2.5 |
| | | 0.5 | 8 | 31.0 ± 3.5 |
| | | 2.5 | 15 | 44.0 ± 5.3 |
| MW3 (Glass wool) | 10.2 | - | - | 0.0 |
| | | 6 | 8 | 7.0±5.3 |
| | | 10 | 9 | 7.0±1.8 |
| | 40.8 | - | - | 0.0 |
| | | 6 | 10 | 11.0±3.5 |
| | | 10 | 15 | 17.0±1.8 |
| | 342.5 | - | - | 2.5 |
| | | 1.1 | 8 | 13.0±3.5 |
| | | 2.3 | 15 | 22.0±12.3 |

### 3.3.2. Temporal variation of fibres in a residential house

In the recently built residential house MMVF surface density began to decrease after two months of completion of construction work in all investigated rooms (**Table 3.8**). In three months after the construction work, MMVF surface density in rooms decreased from 2.3–5.6 to 1.1–2.3 fibre/cm$^2$. After four months in two of five rooms MMVFs were below the detection limit, which is already sufficient to not take any corrective actions. The decrease of MMVFs in ambient air could be determined by the fact that indoor environment was carefully cleaned by the vacuum cleaner every time before exposure of Petri dishes. The lowest fibre surface density in the room No. 3 has been affected by low movement of occupants in this particular area. In case of irritation symptoms for the inhabitants, we recommend the investigation of settled dust to potentially determine if MMVF concentrations are not elevated. Study performed by Palomaki et al. (2008) revealed similar findings that intense cleaning of indoor surfaces should minimize the amount of mineral fibres.

**Table 3.8.** MMVFs surface density after the completion of construction works

| Collection time | MMVFs surface density, fibre/cm$^2$ | | | | | |
|---|---|---|---|---|---|---|
| | Room No. 1 | Room No. 2 | Room No. 3 | Room No. 4 | Room No. 5 | Guideline in Finland |
| After 1 month | 3.4 | 4.5 | 2.3 | 3.4 | 5.6 | |
| After 2 months | 5.6 | 3.4 | 2.3 | 4.5 | 5.6 | 0.2 fibre/cm$^2$ |
| After 3 months | 2.3 | 1.1 | 1.1 | 2.3 | 1.1 | |
| After 4 months | 0.0 | 1.1 | 1.1 | 1.1 | 0.0 | |

### 3.3.3. The presence of mineral fibres in multifamily residential buildings

MMVFs were detected in all ten investigated multifamily buildings, although in relatively low concentrations. Surface density of MMVFs varied in the range of 0.11–0.67 fibre/cm$^2$ (**Table 3.9**). The mean surface density of MMVFs observed in our study was in comparable range to Finnish studies performed by Salonen et al. (2009) (<0.1−5.0 fibre/cm$^2$) and Kovanen et al. (2007) (<0.1–2.6 fibre/cm$^2$). Obtained data was similar with a guideline value of below 0.2 fibre/cm$^2$ (regularly cleaned surfaces), presented in Schneider (2000) and Kovanen et al. (2007) studies. Guideline value of 0.2 fibre/cm$^2$ will be official in Finland from year 2015. A guideline from Finland was selected for comparison because in this case the fibre surface density is used as a main unit not a number of fibres per volume (fibre/cm$^3$).

In general, such low concentrations of mineral fibres do not call for any corrective actions. These results serve as the baseline data for the further monitoring of MMVFs in these buildings after the refurbishment.

**Table 3.9.** Surface density of MMVFs in ten multifamily buildings

| Building No. | 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 | 9 | 10 |
|---|---|---|---|---|---|---|---|---|---|---|
| Fibre surface density, fibre/cm$^2$ | 0.11 ±0.05 | 0.11 ±0.08 | 0.56 ±0.22 | 0.45 ±0.20 | 0.45 ±0.18 | 0.11 ±0.05 | 0.11 ±0.06 | 0.22 ±0.11 | 0.11 ±0.06 | 0.67 ±0.25 |

## 3.4. Spatial and temporal variations of particulate matter concentrations in multifamily apartment buildings during operation phase

A comprehensive assessment of PM concentrations and its variations in Lithuanian multifamily apartment buildings were reported. This part of the study was researched in a framework of INSULAtE ("Improving energy efficiency of housing stock: impacts on indoor environmental quality and public health in Europe") project. The results presented in this chapter reflect a pre–refurbishment measurement phase. Aim of this part was to select comparative parameters and indicators for demonstration of the effects of refurbishment on IAQ: PM temporal variation, PM concentration decay rate, I/O ratio, background concentration and PM fraction ratio.

### 3.4.1. Spatial variation of particulate matter concentrations

*Average particulate matter concentrations in buildings*

In general, the observed levels of PM concentrations varied greatly among the investigated buildings (B#) and apartments (A#). **Table 3.10** summarizes the results of indoor and outdoor PM$_1$, PM$_{2.5}$, and PM$_{10}$ measurements averaged across all buildings.

The highest mean indoor PM$_1$, PM$_{2.5}$, and PM$_{10}$ concentrations were registered in B4 (14.7±5.5 μg/m$^3$), B4 (18.1±6.4 μg/m$^3$), and B10 (38.4±30.0 μg/m$^3$), respectively. Outdoor mass concentrations were highest in B4 for all PM fractions: PM$_1$ (24.2±5.6 μg/m$^3$), PM$_{2.5}$ (37.0±10.4 μg/m$^3$) and PM$_{10}$ (55.8±17.6 μg/m$^3$). The increases in PM levels in B4 may be attributed to the fact that during the week of sampling outdoor temperature was lower (-6 °C) in comparison with other measurement campaigns. Lower outdoor temperature resulted in a more intense fuel burning in the surrounding single–family homes and larger influence of pollution from street traffic due to the decreased atmospheric mixing conditions. In this case, the penetration of PM into indoor environment resulted in higher levels of PM indoors. PM$_{10}$ values measured in B4 exceeded the 24 hour EU air quality standard (50 μg/m$^3$) (WHO 2005).

The average indoor and outdoor PM concentrations observed in this study were compared with those reported in various studies around the world (**Table 3.11**).

Measurements in some studies listed in **Table 3.11** were performed using filter–based techniques, thus providing more accurate estimates of PM levels.

The results on indoor air quality from our study, represented by the $PM_{2.5}$ concentration, are of comparable range with Jung et al. (2010) (New York, $17.8\pm14.9$ μg/m$^3$) and Wichmann et al. (2010) (Stockholm, $8.4\pm3.0$ μg/m$^3$), but significantly lower compared to the recent data from Cao et al. (2012) (Guangzhou, $83.0\pm29.1$ μg/m$^3$) or Massey et al. (2012) (Agra, $146.0\pm39.0$ μg/m$^3$). At the same time, the outdoor PM concentrations were similar to those concentrations measured in I/O campaigns by Diapouli et al. (2011) ($PM_2$ – $31.8\pm7.8$ μg/m$^3$ during cold period) in Athens and Wichmann et al. (2010) ($PM_{2.5}$ – $8.9\pm4.1$ μg/m$^3$ during winter period) in Stockholm. The range of outdoor $PM_{2.5}$ concentrations varied between $6.3\pm2.9$ μg/m$^3$ (B9) and $37.0\pm10.4$ μg/m$^3$ (B4) meanwhile outdoor $PM_{10}$ concentrations ranged between $12.7\pm7.0$ μg/m$^3$ (B5) and $55.8\pm17.6$ μg/m$^3$ (B4).

Several major factors may be attributed to the relatively large dispersion of results between different compared studies. Primarily, the concentration of aerosol particles is influenced by geographical location; weather conditions is important as well; urbanization level of the city in which measurements were performed; abundance of the traffic; distribution of industrial facilities, etc.

**Table 3.10.** Descriptive statistics of indoor and outdoor PM$_1$, PM$_{2.5}$, and PM$_{10}$ mass (μg/m$^3$) concentration measurements. The number in the parentheses (B#) represents building number where the min or max value occurred

| | | Mean ± StDev | CV | Geo. Mean | Median | Percentile 5.0 | Percentile 95.0 |
|---|---|---|---|---|---|---|---|
| **Indoor** | **PM$_1$** | 3.1±3.8 (B9) | 0.33 (B1) | 1.9 (B9) | 1.7 (B9) | 0.9 (B9) | 5.7 (B2) |
| | | 14.7±5.5 (B4) | 0.87 (B8) | 13.6 (B4) | 14.0 (B4) | 7.2 (B4) | 32.6 (B10) |
| | **PM$_{2.5}$** | 4.5±5.3 (B9) | 0.36 (B1) | 2.9 (B9) | 2.6 (B9) | 1.2 (B8) | 8.8 (B2) |
| | | 18.1±6.4 (B4) | 1.07 (B8) | 16.9 (B4) | 17.9 (B4) | 8.7 (B4) | 44.3 (B10) |
| | **PM$_{10}$** | 13.6±12.9 (B5) | 0.42 (B4) | 9.7 (B5) | 9.3 (B9) | 1.8 (B8) | 30.4 (B6) |
| | | 38.4±30.0 (B10) | 1.47 (B8) | 28.1 (B10) | 30.0 (B10) | 10.5 (B4) | 104.8 (B8) |
| | | Mean ± StDev | CV | Geo. Mean | Median | Percentile 5.0 | Percentile 95.0 |
| **Outdoor** | **PM$_1$** | 4.4±2.3 (B9) | 0.23 (B4) | 3.9 (B9) | 3.8 (B9) | 2.0 (B9) | 9.1 (B9) |
| | | 24.2±5.6 (B4) | 0.62 (B6) | 23.6 (B4) | 23.2 (B4) | 16.8 (B4) | 41.1 (B3) |
| | **PM$_{2.5}$** | 6.3±2.9 (B9) | 0.25 (B1) | 5.7 (B9) | 5.6 (B9) | 3.1 (B9) | 11.7 (B9) |
| | | 37.0±10.4 (B4) | 0.61 (B6) | 35.6 (B4) | 35.0 (B4) | 24.2 (B4) | 56.7 (B4) |
| | **PM$_{10}$** | 12.7±7.0 (B5) | 0.27 (B1) | 11.0 (B5) | 11.1 (B5) | 5.1 (B5) | 25.4 (B5) |
| | | 55.8±17.6 (B4) | 1.00 (B8) | 53.1 (B4) | 53.8 (B4) | 32.9 (B4) | 86.3 (B4) |

**Table 3.11.** Comparison of mean measured indoor and outdoor PM concentrations with various studies around the world

| References | PM fraction | Mean Indoor | Mean Outdoor | I/O | Site | City |
|---|---|---|---|---|---|---|
| **Our study** | PM$_1$, PM$_{2.5}$, PM$_{10}$ | 7.3±4.4, 9.8±6.3, 20.8±18.5 | 12.8±5.1, 17.6±7.4, 29.9±15.3 | 0.69, 0.70, 0.98 | Residential multifamily homes | Kaunas, Lithuania |
| **Cao et al. (2012)*** | PM$_{2.5}$ | 83.0±29.1 | 119.9±59.1 | 0.69 | Residential single family homes | Guangzhou, China |
| **Massey et al. (2012)**** | PM$_1$, PM$_{2.5}$, PM$_{10}$ | 135.0±28.0, 146.0±39.0, 254.0±56.0 | 134.0±22.0, 157.0±26.0, 235.0±61.0 | 1.01, 0.93, 1.08 | Residential single family homes | Agra, India |
| **Diapouli et al. (2011)*** | PM$_2$, PM$_{10}$ | 27.2±3.6, 31.8±7.8 | 40.6±24.7, 53.9±18.4 | 0.67, 0.59 | Residential multifamily homes | Athens, Greece |
| **Jung et al. (2010)*** | PM$_{2.5}$ | 17.8±14.9 | 10.9±3.13 | 1.63 | Residential multifamily homes | New York, USA |
| **Wichmann et al. (2010)*** | PM$_{2.5}$ | 8.4±3.0 | 10.3±4.2 | 0.87 | Residential multifamily homes | Stockholm, Sweden |
| **Stranger et al. (2009)*** | PM$_1$, PM$_{2.5}$, PM$_{10}$ | 31.0±12.0, 36.0±13.0, 39.0±15.0 | 29.0±13.0, 36.0±14.0, 41.0±16.0 | 2.0, 1.5, 1.3 | Residential single family homes | Antwerp, Belgium |

\* - measured by filter-based technique
\*\* - measured by OPC

### 3.4.2. Temporal variation of particulate matter concentrations

Median 24 hour's coefficients of variance (CV) for indoor $PM_1$, $PM_{2.5}$ and $PM_{10}$ were 0.48, 0.48 and 0.72, respectively (**Annex 5**). Corresponding values for outdoors were 0.49, 0.44 and 0.43, respectively. Indoor PM was characterized by a higher temporal variation, especially for larger particles, due to proximity of measurement points to indoor pollution sources. Comparable findings were observed by Kearney et al. (2011) (the daily-averaged data ranged from 0.22–0.29 for outdoor and 0.44–0.46 for indoor UFPs) and Wichmann et al. (2010) (CV for $PM_{2.5}$ was 0.15 and 0.07, respectively for indoor and outdoor samples). In addition, Wheeler et al. (2011) noticed higher CV values during winter period than during summer period (0.44 in summer and 0.68 in winter). The highest indoor CVs were observed in B9A5 ($PM_1$ and $PM_{2.5}$) and B8A4 ($PM_{10}$). In these apartments, strong peak pollution episodes were registered, caused by cooking and home maintenance activities (CV >1). These activities have generated substantial amounts of fine particles ($PM_1/PM_{10}$ ratio at peaks 0.48 and 0.72), and also resulted in a fast particle concentration decay rate (>0.5 $h^{-1}$), as summarised in **Table 3.12**. The lowest observed CV value for $PM_1$ occurred in B2A2, occupied by a single elderly inhabitant, with no specific pollution episodes observed. However, moderate PM levels and a rather high overall $PM_1/PM_{10}$ ratio (0.48), together with a low I/O ratio (0.38 for $PM_{10}$) and a slow decay of PM concentration (0.28 $h^{-1}$), may altogether indicate contribution of outdoor particles (**Table 3.12**).

**Table 3.12.** Qualitative characterization of the influence of various pollution sources to the indoor PM indicators

| Source of pollution | Temporal variation ($CV_{24-hour}$) | PM concentration decay rate, $h^{-1}$ | I/O ratio | Nighttime conc., $\mu g/m^3$ | $PM_1/PM_{10}$ |
|---|---|---|---|---|---|
| **Outdoor pollution** | <0.75 (Low) | <0.3 (Low) | <0.5 (Low) | >8 (High) | >0.5 (High) |
| **Indoor smoking** | >0.9 (High) | >1.4 (High) | >0.9 (High) | >10 (High) | >0.35 (High) |
| **Single event sources (Cooking)** | >1.0 (High) | >1.4 (High) | >0.8 (High) | <5 (Low) | >0.45 (High) |
| **Continuous resuspension** | <0.75 (Low) | >1.4 (High) | >0.8 (High) | <7 (Low) | <0.35 (Low) |

### 3.4.3. Particulate matter concentration decay rates

Particle concentration decay rates ($PM_{2.5}$, $h^{-1}$) in ascending order from all apartments are summarized in **Figure 3.6**. Median indoor $PM_{2.5}$ concentration decay rate was equal to 0.32 $h^{-1}$, and varied in the range from 0.14 to 1.93 $h^{-1}$ for B1A3 and B10A4, respectively.

The most extreme cases of a rapid PM concentration decay were associated with smoking indoors (B8A1 and B10A4), when the concentrations increased and decreased sharply relative to each occurrence of smoking. These apartments also revealed elevated concentrations of PM, relatively low I/O values, and high ratios of $PM_1/PM_{10}$ (**Table 3.12**).
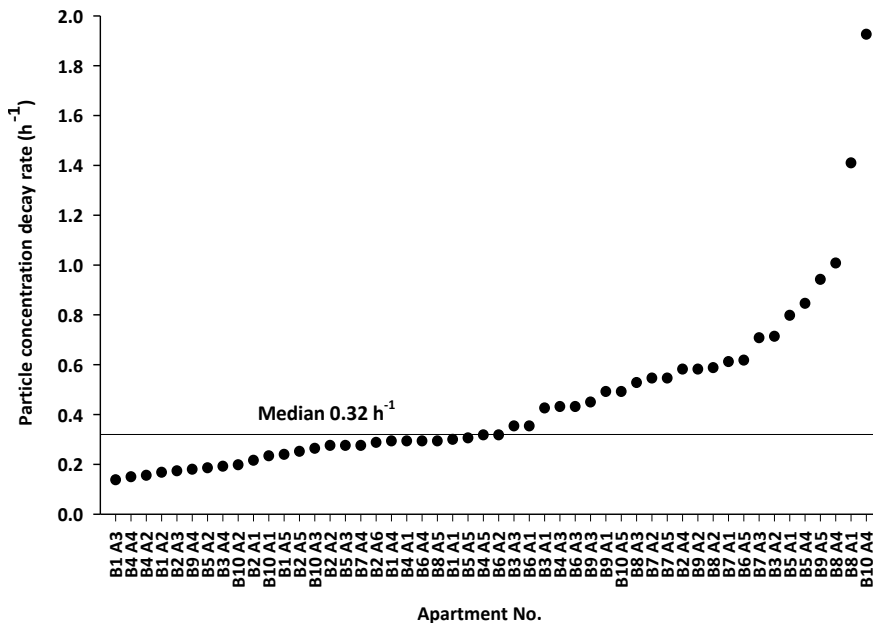


**Figure 3.6.** $PM_{2.5}$ concentration decay rates in ten multifamily buildings ($h^{-1}$)

The lowest decay rates ($<0.2$ $h^{-1}$) were observed in several apartments (e.g. B1A3 and B4A4). These apartments were characterized by moderate PM concentrations, low temporal variation (CV $<$ 0.33 for $PM_{2.5}$), low to average background levels, and I/O ratios mostly below 0.6. The low temporal variation resulted in absence of high peak concentrations. Hence, decay was slow, considering minimal air movement in winter at natural ventilation conditions.

In general, the relationship between air change rate (ACH) and particle concentration decay rate (**Figure 3.7**) was weak and statistically insignificant ($r_S=-0.22$, $p > 0.05$), although the tendency of a reverse relationship was obvious (slope of the regression curve equal to -0.23). Such relationship is expected, as air change

rate is one of the governing factors influencing the removal of fine particles from indoor air. Higher values of the coefficient of correlation might have been obtained in case of parallel measurements of ventilation rate to PM concentration. In this study, we have not aimed for such measurements considering the nuisance for the inhabitants.
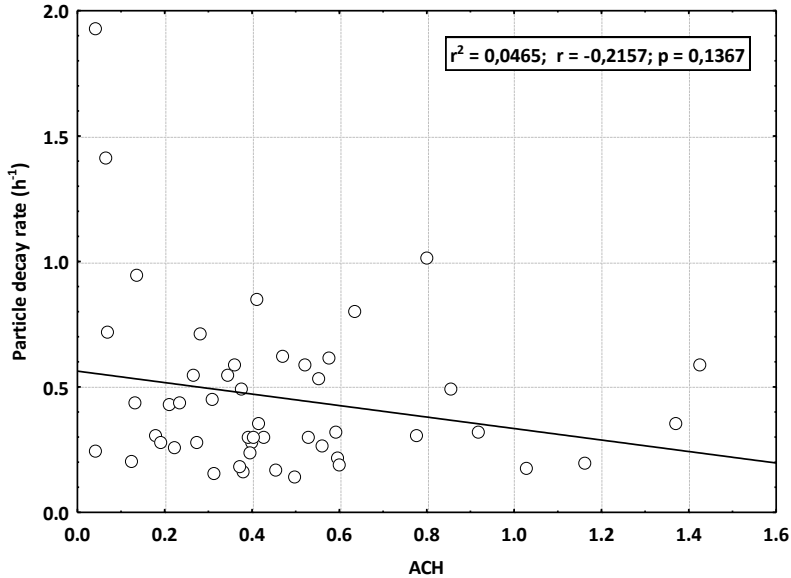


**Figure 3.7.** The relationship between $PM_{2.5}$ concentration decay rate and ventilation rate

The concentration decay rates obtained in our study were at comparable level with ones registered by Özkaynak et al. (1996) – 0.39 $h^{-1}$. At the same time, Long et al. (2001) found even lower decay rates during winter periods (0.10±0.03 $h^{-1}$).

We anticipate that PM concentration decay rate may potentially be the most representative indicator of the effects of building refurbishment on indoor PM concentration. The refurbishment process in Lithuania, similarly to other Nordic countries, usually involves installation of new layers of thermal insulation and replacing old windows. These changes may also result in a more airtight building envelope, which in turn may change pollutant concentration decay patterns due to different removal by ventilation and supply of pollutants from outdoor air.

### 3.4.4. Indicative parameters of particulate matter characterization

*Representation of PSD by ratios of fractions*

$PM_1/PM_{10}$ and $PM_{2.5}/PM_{10}$ ratios for both indoor and outdoor measurements are summarized in **Annex 6**. Indoor $PM_1/PM_{10}$ ratios varied from 0.10 (B9A1) to 0.78

(B4A1), whereas $PM_{2.5}/PM_{10}$ ratios varied from 0.19 (B9A1) to 0.91 (B4A1). Corresponding outdoor $PM_1/PM_{10}$ ratios varied from 0.22 (B9A5) to 0.75 (B3A3) and $PM_{2.5}/PM_{10}$ ratios from 0.34 (B9A5) to 0.91 (B3A3). Low ratios indicate the absence of fine–particle generating sources or the presence of strong sources generating larger particles (such as resuspension of dust during vacuuming) (Chen and Hildemann 2009; Ferro et al. 2004). At the same time, high ratios indicate the presence of fine aerosol generation activities, mainly due to thermal treatment of organics (cooking, candle burning), or penetration of traffic aerosol to indoor environment, especially if no major indoor sources are present. These ratios are useful for determining the source of particles, but they may not be useful for the comparison of the IAQ before and after building refurbishment, since they are very source-dependent.

### *Indoor–to–Outdoor ratios*

The average values for I/O ratios are summarized in **Figure 3.8** and **Annex 7**. Seventy–five percent of $PM_1$ I/O ratios were significantly different ($p < 0.05$) from unity (34 apartments of 45), 80 % of $PM_{2.5}$ (36 apartments of 45), and 51 % of $PM_{10}$ (23 apartments of 45).
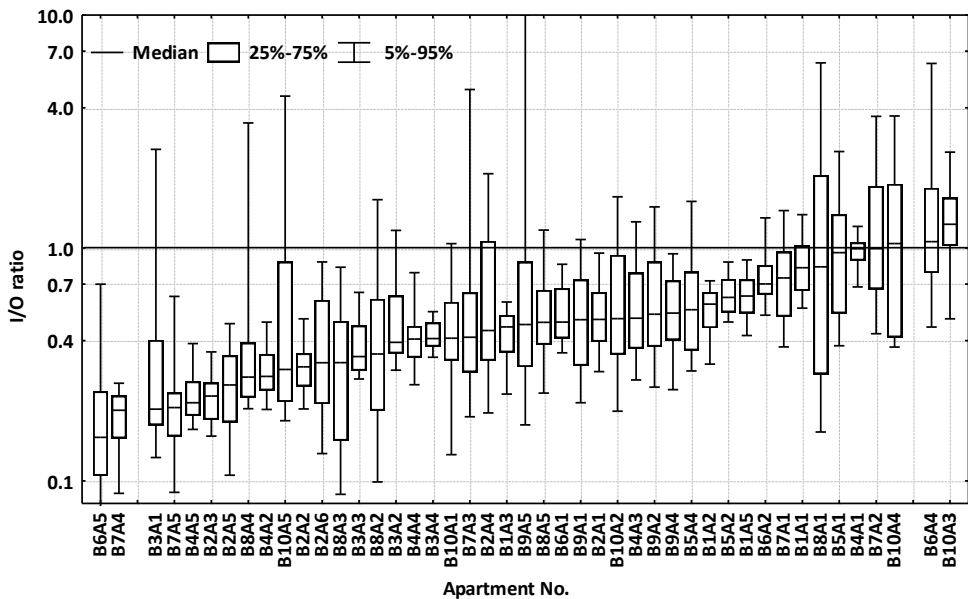


**Figure 3.8.** I/O ratios of $PM_{2.5}$

Maximum median I/O values were registered in B10A3 (1.33 for $PM_1$, 1.26 for $PM_{2.5}$), which could be influenced by intense use of dehumidifier during entire day, as noted by the residents. We assume that the dehumidifier increased PM concentration as a local ventilation device by continuous resuspension when

circulating air within the room. Minimum values were registered in B6A5 (0.19 for $PM_1$ and 0.15 for $PM_{2.5}$), which apartment was unoccupied during the measurement session. Outdoor temperature was rather low, associated with increased outdoor particle concentrations, which in turn caused very low I/O ratios. Average (median) I/O ratio values for Lithuanian apartments were 0.69 in case of $PM_1$, 0.70 for $PM_{2.5}$ and 0.98 for $PM_{10}$.

The observed $PM_{2.5}$ I/O ratios are somewhat lower as compared to similar studies performed in Helsinki, 0.77 (Koistinen et al. 2004), Hong Kong, 0.80 (Ho et al. 2004) and Stockholm, 0.93 (Wichmann et al. 2010). This may indicate stronger presence of fine outdoor particles in Lithuanian apartment buildings due to less airtight building envelopes. I/O ratios of fine particles were lower than that of coarse particles due to a more intense generation of course dust in indoor environment and the proximity of coarse particles to the samplers as opposed to the outdoor particles (Morawska et al. 2001; Poupard et al. 2005). We also did not register a relationship between ventilation rate and I/O ratio ($r_S = -0.02$, $p > 0.05$, slope $= -0.02$), proving that other factors are more important in determining this ratio than the ventilation rate (**Figure 3.9**).
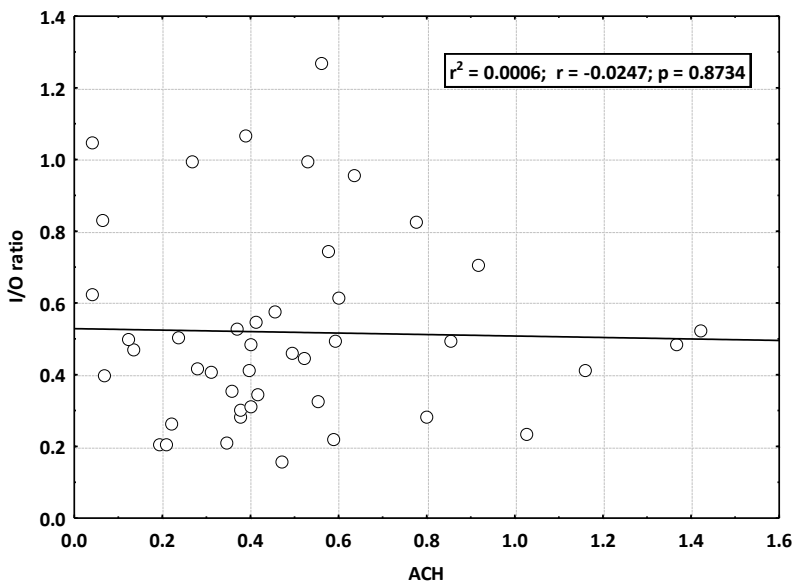


**Figure 3.9.** The relationship between I/O ratios of $PM_{2.5}$ and ventilation rate

*Background (nighttime) particulate matter concentration levels*

Background (nighttime) PM levels were assessed in order to represent background indoor pollution by PM. The background levels represent the lowest contribution of indoor sources to indoor PM concentrations. **Figure 3.10** shows

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$PM_{2.5}$ concentrations (two hour interval of the lowest concentrations) inside of each apartment (the nighttime concentrations of $PM_{10}$ are presented in **Annex 8**). In all cases, the selected nighttime intervals were significantly lower ($p < 0.05$) compared to daytime concentrations. Kearney et al. (2011) reported that the nighttime period of 2–6 am was generally the period of lowest ultrafine particle levels in homes. In this case, this period varied from 2 am to 7 am (sometimes until 8 am), depending on the age of the inhabitants and their daily schedule. These nighttime concentrations of PM represent the combined effects of gravitational and electrostatic forces as well as ventilation flow during the penetration of PM through building envelope cracks and leaky windows.
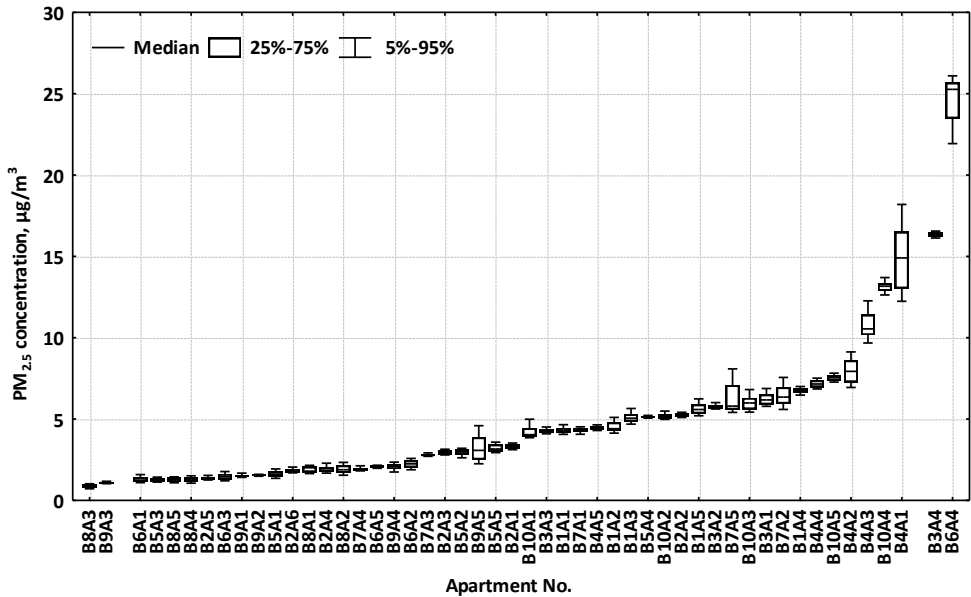


**Figure 3.10.** Nighttime (background) $PM_{2.5}$ levels

Average (median) values of background $PM_{2.5}$ and $PM_{10}$ were 5.0±2.8 and 6.7±3.5 µg/m$^3$, respectively. Only in 10 % of the apartments did nighttime $PM_{2.5}$ exceed 10 µg/m$^3$. The highest background $PM_{2.5}$ was recorded in B6A4 (24.6 µg/m$^3$). Occasional smoking indoors and use of air dehumidifier during night may have been the main reason for such high nighttime PM concentration level in this apartment. This effect may be characterized by high average PM concentration, high I/O ratio, relatively low $PM_1/PM_{10}$ ratio, but at the same time low temporal CV and low concentration decay (**Table 3.12**). This is different from purely intensive smoking, which may be characterized by fast concentration decay, high temporal CV and relatively high $PM_1/PM_{10}$ ratio.

Building 4 also stood out with relatively high indoor PM concentrations ($PM_{10}$ – 11.3±5.7 µg/m$^3$). The measurements in this building corresponded to the increased outdoor pollution event, thus it can be presumed that the influence of outdoor

pollutants to IAQ was registered. The apartments in B4 were characterized by the highest $PM_1/PM_{10}$ ratio (0.65), slow particle concentration decay (<0.45 $h^{-1}$), and relatively high background concentration values (**Table 3.12**).

The analysis of the effect of ventilation rate to the background aerosol concentration revealed a similar type of a weak adverse relationship ($r_S = -0.08$, $p > 0.05$, slope of the regression curve -1.13) to PM concentration decay rate (**Figure 3.11**). It was hypothesized that the increasing ventilation rate should decrease nighttime particle concentrations. It is obvious that this relationship is largely affected the presence of strong indoor pollution source, such as smoking (B10A4) or particle resuspension by local ventilator operating throughout the nighttime (B6A4). Having these cases removed from the correlation analysis, the $r_S$ value was equal to -0.003.
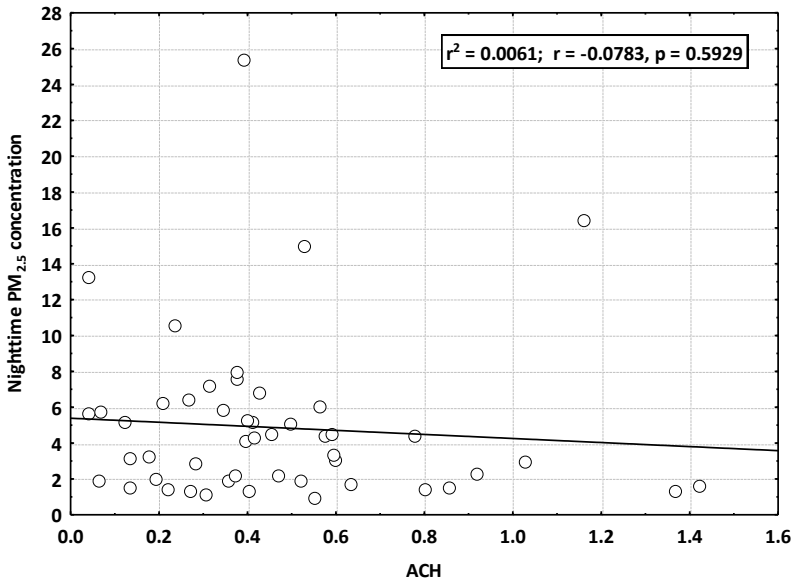


**Figure 3.11.** The effect of ventilation rate to the background aerosol concentration ($PM_{2.5}$)

We anticipate that the building refurbishment may affect the background concentration levels of PM, but the effect may be non–uniform. Nighttime usually excludes effects of human activities, so the resulting concentration could be considered representative of the true pollution of the living space. Increased air tightness of the building envelope may decrease the influence of outdoor particles on the overall balance of PM concentrations. On the other hand, decreased natural ventilation may keep the particles elevated for longer periods. This potential indicator should be coupled with particle concentration decay rate as well as I/O ratio and others.

### 3.4.5. Indoor microclimate parameters and building information

Concentrations of measured environment parameters, including $CO_2$, CO, T and RH in the coldest spot (Tc and RHc) and warm area (Tw and RHw) are presented in **Table 3.13**.

30% of investigated apartments had 24-hour average $CO_2$ concentrations higher than 1200 ppm; and 41 % apartments were above 1000 ppm. Considering 8-hour maximum values, 31 % apartments had levels $\geq$1200 ppm. $CO_2$ concentrations during occupied periods were considerably elevated. Twenty eight apartments had low CO levels below the national threshold value (2.43 ppm in 24 hours).

Tw was observed lower than 20 ℃ in 61 % apartments (11 % below 18 ℃, lowest equal to 14.4 ℃). RHw in 36 % apartments were below 40 %, and 6 % exceeded 60 % (highest equal to 69.5 %). 54 % of apartments had lower than recommended temperature (Tw < 20 ℃) during the measurement period.

T and RH in the coldest spots and warm areas were significantly correlated, e.g., Tc and Tw had Spearman correlation coefficients of 0.73. The correlation coefficients between indoor and outdoor T (To) (Tc and To, Tw and To) were 0.60 and 0.42, respectively. To and RHo data during the corresponding PM measurement period were obtained from Kaunas meteorological monitoring station and are summarized in **Annex 9**.

**Table 3.13.** Indoor and outdoor $CO_2$, CO, temperature and relative humidity in the coldest spot (Tc and RHc) and warm area (Tw and RHw) (Table shows average, standard deviation, median, and 95th percentile)

| Parameter | Unit | Average | SD | Median | 95th |
|:---:|:---:|:---:|:---:|:---:|:---:|
| Tc | °C | 17.9 | 2.2 | 18.3 | 21.0 |
| Tw | °C | 19.6 | 1.6 | 19.6 | 22.2 |
| RHc | % | 48.6 | 10.6 | 47.4 | 68.6 |
| RHw | % | 43.5 | 9.9 | 43.4 | 63.8 |
| $CO_2$ | ppm | 1027 | 386 | 968 | 1807 |
| CO | ppm | 0.16 | 0.46 | 0.00 | 1.34 |

The effect of building refurbishment in the tested apartments revealed non uniform results with respect to thermal conditions. The representative long-term variations of temperature and RH in three selected apartments are presented in **Figure 3.12**, showing data from the 14 months of measurements. During the first heating season, the indoor temperatures were kept in the interval of 14-20 degrees, while RH varied between 30-70 %. The refurbishment occurred during September-December 2012, and involved installation of external wall insulation, replacement of windows and heating systems of the building. It is evident that the thermal comfort has increased, with the temperatures ranging between 19-22 °C and RH at 40-60 %

during the heating season. It is also evident, that orientation of the apartment (south/north) played major role in the levels of temperature and RH.
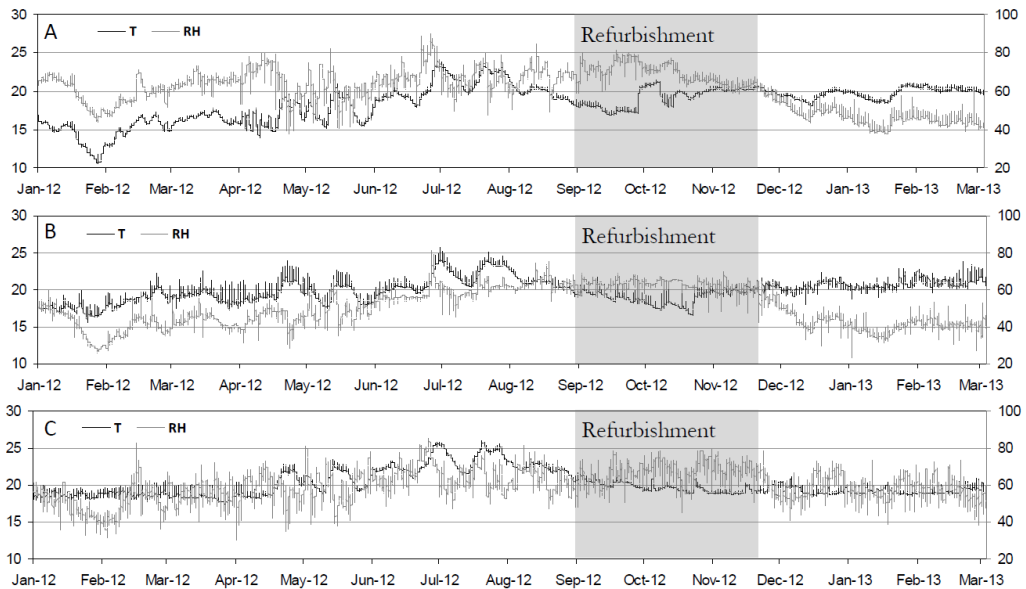


**Figure 3.12.** Variation of indoor temperature and relative humidity in three apartments of a multifamily building in Kaunas, Lithuania (A – north oriented, B and C – south oriented apartments)

Building air-tightness together with ventilation effectiveness are major parameters influencing PM exposure levels and pathways indoors. Focusing only on assurance of more airtight dwellings during refurbishment processes may increase the exposure to indoor air pollutants, resulting in adverse health effects, therefore it is beneficial to understand how building refurbishment changes the air-tightness of the building. Data related to the adaptation and upgrading of the existing buildings was introduced in an extensive report (European Commission 2013). The four-storey apartment building and sports hall were selected as the research objects where air-tightness was measured before and after the refurbishment. The original air-tightness (n50) of the four-storey apartment building was 3.0 $h^{-1}$. After the refurbishment, the air-tightness was observed about 1.0 $h^{-1}$. The air-tightness test was carried out before the refurbishment in the sports hall as well, but it failed because the building was too leaky. After refurbishment IR surveys and an air-tightness test were performed again. The air-tightness test demonstrated good air-tightness with an n50-value of 0.63 $h^{-1}$. It is evident that refurbishment of buildings impacts air-tightness significantly, therefore only tightening the building envelope and this way reducing natural ventilation may decline the indoor air exfiltration outdoors and increase the residence time of contaminants indoors.

Replacement of "old windows" with modern double-glazed windows reduces the air infiltration indoors and negatively impacts ventilation levels as well. The

study performed by Oreszczyn (2005) revealed that the installation of modern replacement windows reduced the predicted heating season mean background air change rate by an average of 0.23±0.08 ACH. Similar results were obtained in the Ridley (2003) study, were replacement of old windows reduce background ventilation rate by 0.25 ACH to the average property (0.38 % reduction).

Ventilation may vary considerably with time, especially in naturally ventilated dwellings, due to the variation in the ventilation driving forces as induced by the indoor/outdoor temperature difference and wind pressure. However, the influence of the occupants' behaviour may also be an important factor (Stymne et al. 2006). The effectiveness of natural ventilation was determined by anemometer by measuring air flow velocity through the air exhaust channels (mostly in kitchens). The results showed that about 65 % of apartments did not meet the minimum requirement for an air change rate of 0.50 $h^{-1}$. The median ACH in dwellings was 0.40 $h^{-1}$ (mean value – 0.46 $h^{-1}$). ACH varied greatly among tested apartments. Lowest ACH was observed in B1A5 and B10A4, and equal to 0.04 $h^{-1}$, maximum value of 1.43 ACH was measured in B9A2.

The results obtained from Dimitroulopoulou (2011) review study had showed that the occupants' behaviour (i.e. window opening depending on the season) strongly affects the whole building ventilation as well. Moreover, houses need to be airtight to conserve the warm in the building in colder climate zones. In this case, natural ventilation is often unable to provide adequate ventilation for odour or contaminant removal, therefore mechanical ventilation is necessary to achieve minimum ventilation rates. For this reason the ventilation did not fulfil the minimum requirements in most of monitored dwellings in the Nordic and Eastern Europe countries.

Data gathered from housing questionnaires and diaries revealed substantial information. Response rate of answered questionnaires was 58 %. The respondents were relatively elder people (averaged ~54 years) and a larger percentage was female (64 %); average number of occupants living in the apartment was 2.6. About 42 % occupants had pets (dogs, cats, guinea pigs, birds, etc.) and kept them inside.

Some 59 % of the respondents were satisfied or fairly satisfied with indoor air quality. Most of the respondents (82 %) reported relative low indoor heating temperature (≤20 ºC), 38 % reported too cold temperature.

Respondents reported that gas stove (84 %) was predominant facility in kitchens and 75 % apartments had installed kitchen vent hoods. Daily or almost daily opening of windows varied from 33.3 % (other area) to 72.5 % (bedroom).

Simplified data from housing questionnaires and diaries, designed for gathering the information from the apartments, their occupancy and occupant activities, is presented in **Annex 10**.

### 3.4.6. Comparison of particulate matter concentrations before and after refurbishment in multifamily buildings

The current section describes the comparison of PM concentrations before and after refurbishment in multifamily buildings focusing on several comparative parameters and indicators (I/O ratio, PM nighttime concentration, ratio of PM size fractions, and PM concentration decay rate).

The comparison of PM concentrations ($PM_{2.5}$) was performed based on indoor and outdoor PM concentration levels, I/O ratios, PM background (nighttime) concentrations, ratios of PM size fractions, and $PM_{2.5}$ concentrations decay rates before and after the refurbishment in Lithuanian multifamily buildings.

In general, the observed full day median $PM_{2.5}$ concentration levels indoors and outdoors (before indoor – 8.3, outdoor – 15.9 μg/m$^3$; after indoor – 9.7, outdoor – 20.5 μg/m$^3$) as well as I/O (before – 0.49, after – 0.46) ratios revealed similar numbers before and after the refurbishment (**Figure 3.13 and Figure 3.14**). Overall, the half of apartments (15 apartments from 30) revealed higher indoor $PM_{2.5}$ concentrations after the refurbishment. In case where extreme indoor pollution (smoking) occurred, the difference was more than three times higher (31.2 μg/m$^3$ and 106.5 μg/m$^3$) after the refurbishment. It could be explained by more airtight building envelope and reduction in natural ventilation.
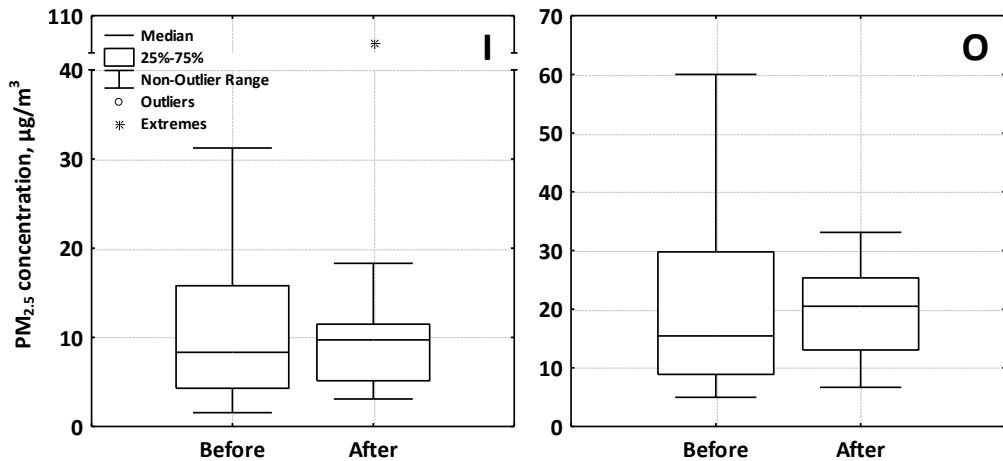


**Figure 3.13.** Comparison of indoor and outdoor $PM_{2.5}$ concentrations before and after refurbishment
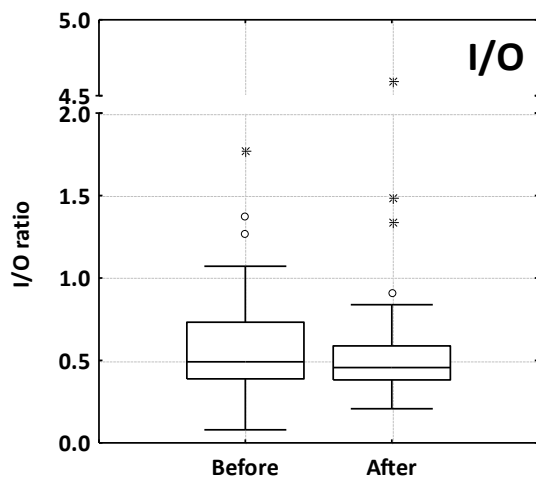
**Figure 3.14.** Comparison of PM$_{2.5}$ I/O ratios before and after refurbishment (I/O – indoor/outdoor ratio)

Background (nighttime) PM$_{2.5}$ concentrations showed comparable trends to full day PM$_{2.5}$ concentration measurements (**Figure 3.15**), i.e. slightly higher concentrations after the refurbishment (18 apartments from 30) and obviously higher concentrations in "smoking" apartment (before – 33.5, after – 75.5 μg/m$^3$).

Median values of nighttime PM$_{2.5}$ I/O ratios were lower (no statistically significant difference) before refurbishment (0.34) compared to ratios observed after refurbishment (0.38) (**Figure 3.16**). However, comparing data obtained from "smoking" apartment it is obvious that the I/O ratio was significantly different (before – 1.33, after – 10.4). Nighttime PM$_{2.5}$ levels, even when pollution sources were no longer active for a few hours, still remained very high due to the lack of natural ventilation indoors. Fine fraction of PM can remain airborne throughout all night under low natural ventilation conditions.

A Wilcoxon signed–rank test was used to test if selected parameters were statistically significantly different before and after refurbishment. The change that occurred with the treatment is not great enough to exclude the possibility that it is due to chance (comparison of indoor conc. – p = 0.869, comparison of outdoor concentration – p = 0.931, and comparison of I/O ratio – p = 0.846).
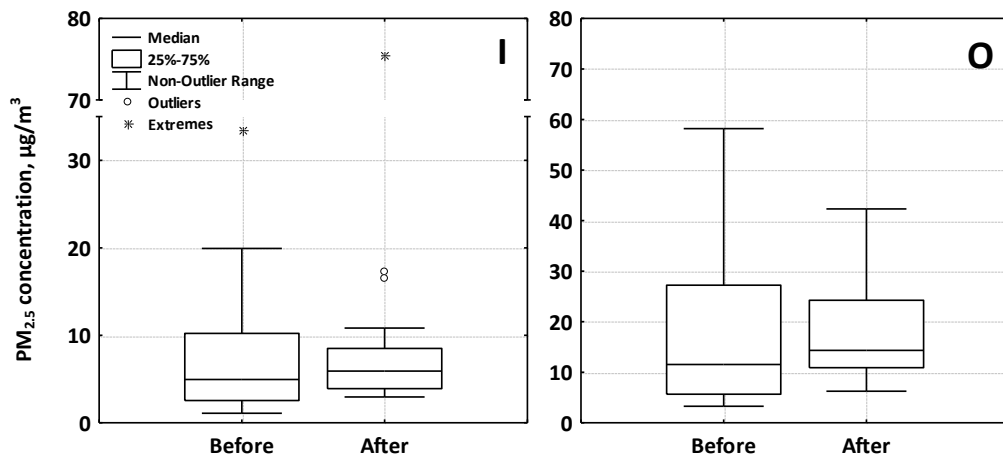
**Figure 3.15.** Comparison of background (nighttime) PM$_{2.5}$ concentrations before and after refurbishment (I – indoor, O – outdoor)
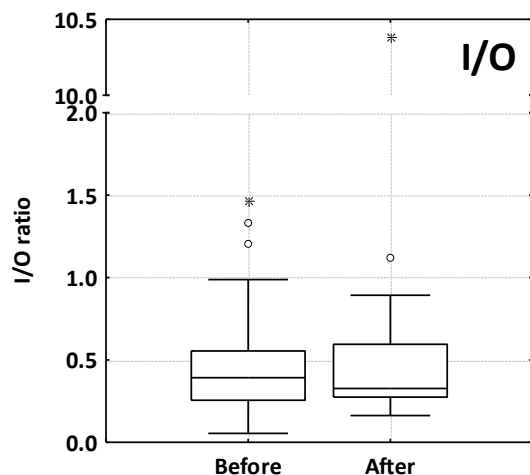


**Figure 3.16.** Comparison of background (nighttime) PM$_{2.5}$ I/O ratios before and after refurbishment

Median indoor PM$_{2.5}$ concentration decay rates were higher before (0.23 h$^{-1}$) than after building refurbishment (0.17 h$^{-1}$) (**Figure 3.17**). Two highest observed PM$_{2.5}$ concentration decay rates (before refurbishment – 1.93 and 1.46 h$^{-1}$) were 3.3 and 4.2 times lower after refurbishment. Overall, particle concentration decay rate decreased by two times. These changes may be influenced by a more airtight building envelope, which in turn may change pollutant concentration decay patterns due to different removal by ventilation and supply of pollutants from outdoor air.

Comparison of indoor and outdoor $PM_{2.5}/PM_{10}$ ratios before and after refurbishment is presented in **Figure 3.18**. Higher median PM fraction ratios were assessed before refurbishment (before – 0.65, after – 0.54), but no statistically significant difference was observed ($p = 0.635$). However, we can assume that decrease of $PM_{2.5}/PM_{10}$ ratio after building refurbishment could be influenced by tighter building envelope, because major fine particle sources are attributed to outdoor pollution (transport and fuel burning emissions).
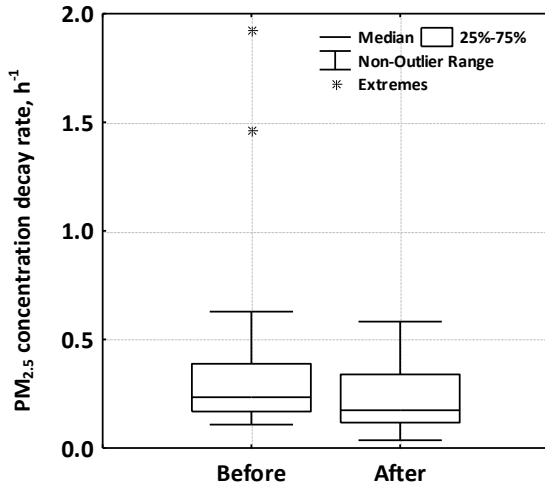


**Figure 3.17.** $PM_{2.5}$ concentration decay rates ($h^{-1}$) before and after refurbishment
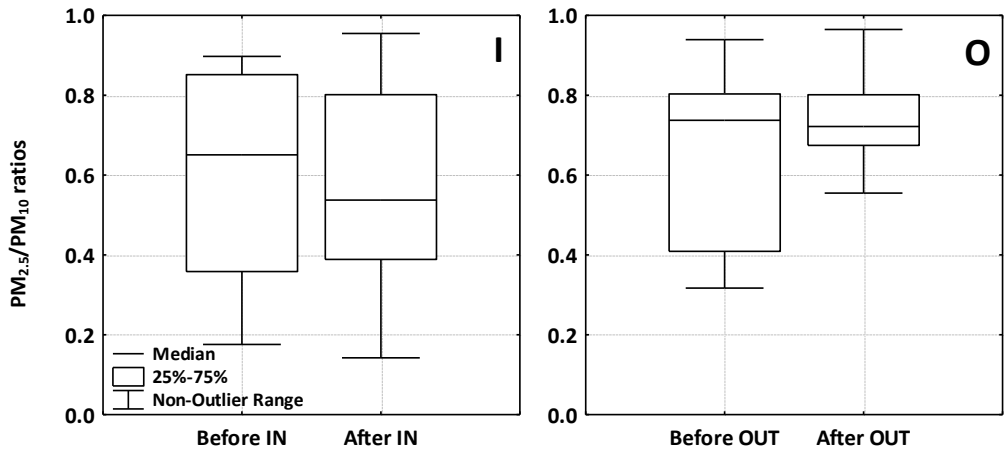


**Figure 3.18.** Indoor and outdoor $PM_{2.5}/PM_{10}$ ratios before and after refurbishment

### 3.4.7. Comparison of minimum and maximum particulate matter concentration values before and after refurbishment

Five minimum (0~20 percentile) and five maximum (~80–100 percentile) $PM_{2.5}$ concentration values were taken with aim to more comprehensively analyse the data obtained before and after refurbishment. Such analysis eliminates the average values and focuses only on extreme cases, e.g. smoking apartment.

Comparison of full day and background (nighttime) indoor $PM_{2.5}$ concentrations, $PM_{2.5}$ I/O ratios, $PM_{2.5}$ concentration decay ratios, and $PM_{2.5}/PM_{10}$ ratios was performed. The Mann–Whitney U test was utilized for the determination of significance of difference of full day and nighttime indicators before and after refurbishment (**Table 3.14**). The value with p level below 0.15 could be identified to be statistically significant different in following comparison.

Minimum median $PM_{2.5}$ concentration values were significantly higher ($p<0.05$) after refurbishment during both full day (2.7 and 8.1 $\mu g/m^3$) and nighttime (1.6 and 5.0 $\mu g/m^3$) measurements (**Figure 3.19**). However, median maximum $PM_{2.5}$ concentration values were observed lower after refurbishment (full day – 19.9 and 12.7 $\mu g/m^3$, nighttime – 19.9 and 9.2 $\mu g/m^3$).

Three times higher minimum median full–day and nighttime $PM_{2.5}$ concentrations could be influenced by two factors: mainly by longer PM residence and reaction time indoors due to tighter building envelope and in some cases due to outdoor pollution influence, because of more frequent opening of windows resulted by higher indoor temperatures after the refurbishment. The opposite scenario (lower values) with maximum median $PM_{2.5}$ concentrations is associated with more frequent opening of windows as well. However in this case the dilution of higher indoor PM concentration with outdoor PM concentration took place.
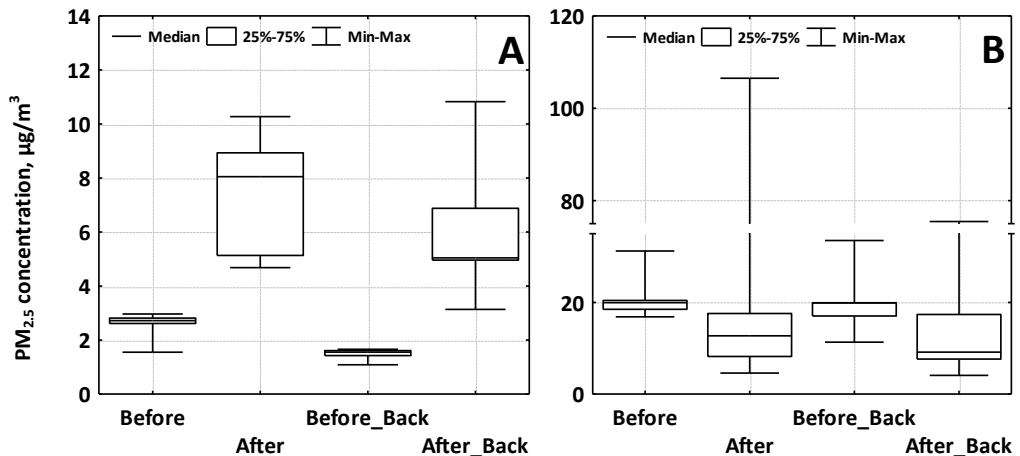


**Figure 3.19.** Comparison of five minimum (A) and maximum (B) $PM_{2.5}$ concentration values before and after refurbishment (Before_Back – background concentration before refurbishment; After_Back - background concentration after refurbishment)

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Comparison of different PM indicators (I/O ratio, PM concentration decay rate and PM size fractions ratio) before and after refurbishment was performed with the apartments where the lowest and highest nighttime $PM_{2.5}$ concentration were observed.

Median minimum nighttime $PM_{2.5}$ I/O ratios were lower (not statistically different, $p = 0.75$) after refurbishment (0.38 and 0.28) (**Figure 3.20**). However, median maximum $PM_{2.5}$ I/O ratios were observed slightly higher after refurbishment (0.44 and 0.48).
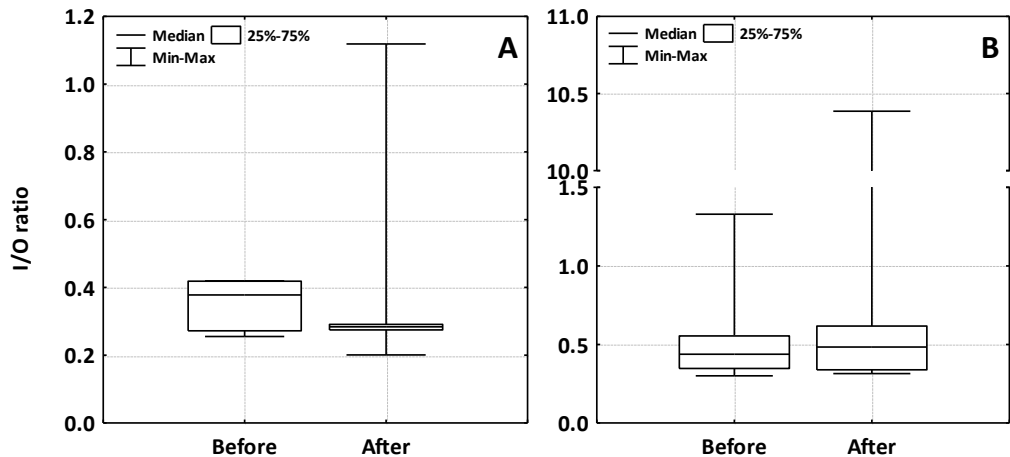


**Figure 3.20.** Comparison of minimum (A) and maximum (B) nighttime $PM_{2.5}$ I/O ratios before and after refurbishment

Median minimum $PM_{2.5}$ concentration decay rates were higher after refurbishment (0.30 and 0.34 $h^{-1}$) and median maximum $PM_{2.5}$ concentration decay rates were lower after refurbishment (0.18 and 0.14 $h^{-1}$) (**Figure 3.21**). No statistically significant differences were observed on both cases (p value equal to 0.92 and 0.46).

After building refurbishment minimum values of $PM_{2.5}$ concentration decay rates were assessed slightly higher possibly because the initial concentration ($C_0$) was elevated thus influencing more rapid concentration decay rates.

Lower values of maximum $PM_{2.5}$ concentration decay rates after refurbishment could be addressed to the decline of air exfiltration and finally increase of indoor PM residence time.
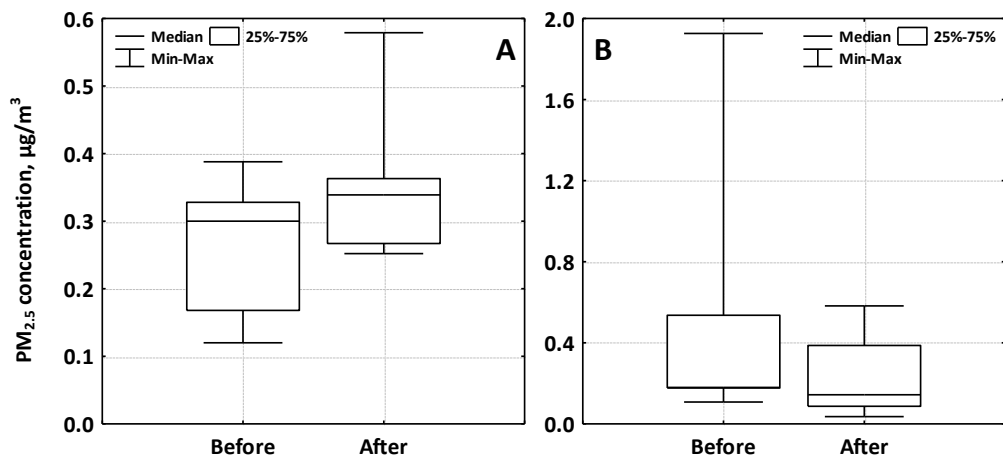
**Figure 3.21.** Comparison of minimum (A) and maximum (B) nighttime PM$_{2.5}$ concentration decay rates (h$^{-1}$) before and after refurbishment

Statistically significant difference (p = 0.12) was observed comparing median minimum nighttime PM$_{2.5}$/PM$_{10}$ ratios (0.44 and 0.70) of corresponding apartments after building refurbishment (**Figure 3.22**). Increased PM$_{2.5}$/PM$_{10}$ ratio of minimum values during the nighttime could be caused by higher residual contamination from cooking or outdoor sources and reduced impact of the natural ventilation. However, maximum nighttime values were not affected by these processes after building renovation (PM$_{2.5}$/PM$_{10}$ ratio of 0.90 and 0.92, p = 0.75).
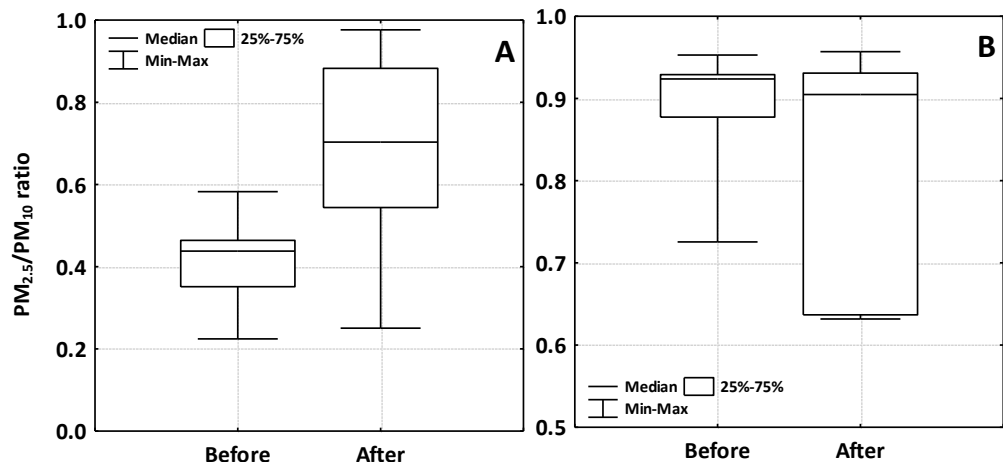


**Figure 3.22.** Comparison of minimum (A) and maximum (B) nighttime PM$_{2.5}$/PM$_{10}$ ratios before and after refurbishment

**Table 3.14.** Comparison of statistically significant difference between minimum and maximum values of selected indicators

| p level < 0.05 | MIN values | MAX values |
|---|---|---|
| PM$_{2.5}$ conc. (Full day) | 0.01 | 0.18 |
| PM$_{2.5}$ conc. (Night) | 0.01 | 0.25 |
| PM$_{2.5}$ I/O ratio (Full day) | 0.75 | 0.10 |
| PM$_{2.5}$ I/O ratio (Night) | 0.75 | 0.75 |
| PM$_{2.5}$ conc. decay rate | 0.92 | 0.46 |
| PM$_{2.5}$/PM$_{10}$ ratio (Full day) | 0.75 | 0.08 |
| PM$_{2.5}$/PM$_{10}$ ratio (Night) | 0.12 | 0.75 |

More comprehensive analysis of lower (0~20) and upper (~80–100) percentiles revealed the statistically significant differences upon several selected indicators before and after refurbishment (**Table 3.14**). The elimination of average values from statistical processing appeared to be a good tool for deeper analysis of the data. It is easier to identify and interpret the changes using following statistical approach.

The analysis identified the overall increase of minimum PM$_{2.5}$ concentration values indoors both during the nighttime and full day observations. This particular case identifies the negative impact of refurbishment processes on IAQ from the PM perspective. Negative effects are mostly associated with the increase of building envelope tightness and eventually decrease of the natural ventilation.

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

## 4.1. Assurance of healthy indoor air quality during building refurbishment and operation

### 4.1.1. Minimization of environmental impact during building construction/ refurbishment

The presented research shows substantial presence of fine aerosol particles (<2.5 μm) in powdery building materials and cement bound materials. Although the dustiness of these materials is a known issue, the impact of these materials to air quality during construction phases should not be underestimated having the new evidence of the particle size distribution and potential health effects. The release of PM should be strictly controlled during construction processes by taking appropriate measures, such as covering of piles of powdery materials against wind resuspension; prevention of the extensive dust aerosolization during the transportation and unloading operations of powdery materials. The advisory measures for worker personal protection should be followed during the construction phase. At the same time, after the construction works are over, a thorough cleaning of all indoor surfaces is recommended. Particles of dust which are retained in indoor environment during the construction/refurbishment phase may be released to indoor air after a substantial period of time after the construction works are over.

The mineral fibre release from asbestos–cement sheet operations has been well documented, and re–confirmed the potential risks associated with the handling of these materials. The obtained emission factors may be used for the estimation of environmental impact as well. The potential hazard of cellulose fibres to human respiratory system is considered as substantially lower than that of asbestos, although having only limited evidence on cellulose fibre adverse effects on the airways. The provided emission factors may further be utilized for the epidemiological studies.

The hazardous properties of MMVFs are believed to be attributable to its fibrous nature rather than to its chemical composition. The most common negative health effects associated with MMVFs is temporary skin, eyes, nose, and throat irritation. However, this material is likely to be and already is widely utilized in the increasing building energy performance. Thus, a substantial presence of MMVFs is expected in indoor environments. Results have shown that there is a risk of continuous fibre penetration to the indoor environment after the construction works are completed. The prevention of fibre pathways through the layers of building envelope should be achieved by sealing joints in and between the ceiling and internal wall assemblies with the barrier materials. The fixtures for lighting should

be carefully selected as they minimize the tightness of the building envelope, both from the energy saving and fibre penetration perspective. The decrease of MMVFs in the ambient air could be achieved by carefully cleaning of indoor surfaces by the vacuum cleaner every time before the exposure of mineral wool insulation.

## 4.1.2. Maintaining healthy indoor air quality in refurbished buildings

Indoor air quality is a very complex phenomenon thus cannot be easily managed. In general, the foremost aim of IAQ management should be concentrated towards assuring suitable microclimate as well as low contamination of indoor air. The main following aspects when assessing refurbished or newly constructed buildings of indoor air quality is suggested:

a)    **The possible sources of pollutants.** This dissertation has focussed on particulate matter as air pollutant. This pollutant has sources in almost every below listed categories. Sources of indoor air contaminants could be categorized into the following major groups:

- Ambient air pollution (fuel combustion, traffic, urban and industrial activities) comes into the building through the ventilation system or by infiltration (building envelope permeability). It was found that outdoor air is the most consistent and important pollutant due to its continuous effect, especially in non–renovated and energy inefficient buildings. Moreover, countries of Northern and Central Europe face extreme outdoor pollution levels due to the increased combustion of fuels for energy production and cold–start transport emissions. The penetration of outdoor PM to the indoor environment contributes from 10 to 60 % of fine fraction of indoor aerosol concentrations. It mainly affects IAQ background levels, i.e., when no indoor pollution sources are present, as well as cause lower decrease rates of PM levels.

- Building materials and furnishings (adhesives, paints, insulation materials, wall and floor coverings, etc.). The emissions of building materials are usually of concern due to emissions of gaseous pollutants, such as volatile organic compounds. The pollutants are continuously emitted in low concentrations and in cases of insufficient ventilation may significantly affect the IAQ. With respect to the aerosol emissions, it has been shown within the research of this dissertation that building thermal insulation may be a significant and continuous source of particles in the air.

- Processes that occur within buildings (combustion, heating, ventilation and air conditioning systems, tobacco smoking, use of cleaning products, cooking, paper processing such photocopying, domestic animals, etc.). Active sources of pollution are the most important contributor to acute increases of air

pollution levels, including aerosol emissions. In these cases, I/O ratio is often increased to the levels of >1.0. Cooking, smoking, and household cleaning products are major sources of particles and gaseous pollutants. Continuous effects of these pollution sources maintain high levels of pollutants and create unhealthy conditions to inhabitants. Moreover, our research indicates that operation of the dehumidifier/humidifier may also contribute to the increased aerosol concentrations due to the resuspension of particles.

- Water and soil (air pollutants coming through water supply, radon and contaminated soils). The hazards of air pollutants are specific to some geographical areas or residential territories erected in former industrial sites. In Lithuania, the hazards from soil radon are low, thus are not of a high concern.

b) **Measures to reducing the levels of airborne particulate matter.** The core measure to reducing any type of pollution is prevention. This is especially valid for the management of IAQ. However, PM is a complex pollutant whose emissions cannot be always avoided. Outdoor PM is difficult to control in urban environments, where traffic pollution and fuels combustion emissions are prevalent. The increased tightness of building envelope contributes to lowering the penetration of outdoor PM. This measure, however, may reduce the ventilation and decrease the removal rate of particles generated indoors. It is also difficult to prevent cooking related emissions. The residents should be warned on the effects of candle burning and incense smouldering as well as the utilization of household cleaning products, as these actions introduce high numbers of nano–sized particles to the indoor air.

It has been discovered that the systems of natural ventilation is not efficient of the rapid dilution of indoor pollutants, as compared to the mechanical ventilation. The introduction of mechanical ventilation in the renovated buildings should be strongly considered as one of the most viable options for the improvement of IAQ. There is a risk, however, that during winter pollution episodes the forced ventilation may introduce outdoor pollutants with higher efficiency, thus proper filtration of outdoor air must be assured. Same applies to the opening of windows for ventilation – this measure is efficient in the areas where outdoor air quality is sufficient to dilute indoor air pollutants, but in the areas of nearby intensive traffic sources or fuel combustion, this may cause an opposite effect.

c) **Assessment of IAQ.** The assessment of IAQ is necessary to monitor the effects of building refurbishment to IAQ. It is suggested that the assessment of IAQ should become a standard procedure in the building energy certification.

The perception of IAQ strongly depends on a single human being, thus there are several options to investigate the IAQ – a subjective and an objective approach. The subjective approach is questionnaire–based. It has been found that the questionnaires as an inexpensive method for the evaluation of IAQ. The questionnaire method was recently investigated as a viable option for the objective IAQ assessment. At the same time, a low degree of participation of the inhabitants was experienced, where only 60 % response rate was achieved. The lack in responsiveness may affect the results of the studies. Moreover, the inhabitants may not interpret all the sources related to the emissions of pollutants as of potential harm to IAQ (especially in cases of aerosol emission).

It was suggested to utilize simplified methods for the monitoring of IAQ, which would include real–time sampling and logging of the IAQ proxies. Among these, new and inexpensive sensors of $CO_2$ and VOCs, as well as passive (diffusion–based) aerosol samplers may provide interesting data with respect to pollution levels and variation. However, the application of such sensors requires further investigations.

## 4.2. Research studies on different indoor aerosol measurements techniques

### 4.2.1. Chamber–based simulation of aerosol emissions

Several experimental approaches for the evaluation of aerosol generation from operations involving powdery building materials, processing of cement–fibre matrix based roof coverings, as well as operation of ceiling structure involving mineral fibre based thermal insulation were suggested in this dissertation. Small–scale chamber methods that were employed in these studies proved itself as an adequate method for the testing of emission factors. The CFD (Computational Fluid Dynamics) modelling is recommended to be executed with the aim to research air flows within the chamber and to select optimal sampling locations.

It was able to simulate both treatment procedures of cement sheet samples, as well as gravitation and fluidization processes of powdery building materials. The key element for an adequate study lies in the selection of analytical equipment to investigate processes with powdery and fibrous aerosols. The measurement of particle number–based emission factors is the most representative (as opposed to the mass–based emission factors). Moreover, particle counters measuring the aerodynamic size of particles (such as time of flight–based or impactor) provide better estimates on particle behaviour in case of inhalation. Moreover, some differences in PSDs measured by optical and aerodynamic methods were registered, indicating that these counters register particles in slightly different ranges based on their morphological properties.

The phase–contrast optical microscopy method was found as a useful tool for the fibre counting, although in some cases, an ordinary light microscopy served as well as phase contrast. An interesting addition to the sampling set–up would include real–time fibre measurement instrument, which would allow monitoring the dynamics and removal of fibre emission.

## 4.2.2. Monitoring of indoor aerosols in multifamily buildings

The monitoring of PM in multifamily buildings provides several major challenges to the researchers. Usually, the studies have to include a relatively large sample of apartments in order to provide stronger results from statistical point of view. This, in turn, raises a challenge of the involvement of equipment and personnel for the sampling campaigns. Particle counters are comparatively expensive equipment (compared to, e.g., temperature or $CO_2$ loggers). At the same time, real–time sampling is important to adequately characterize the dynamics of the processes indoors. That is why the analysis of passive sampling, based on the settled dust measurement, is not informative, and should be utilized for sampling for fibres or microorganisms.

While the choices of methods are broad, it was decided to limit the extent of the assessment, and use active sampling methods due to two reasons: 1) the above presented measures will increase budget of the measurement campaigns, possibly providing only limited information for the objective of the study; 2) The aim of the measurement campaign is cause as little as possible nuisance for the apartment inhabitants. Sampling equipment, operating on an active basis, is causing significant disturbance (e.g. noise) inside of apartments. Our experience shows that this is often a prevailing factor for refusals to participate in the studies. Pumpless diffusion based aerosol samplers (such as nephelometers) may be a viable solution to employ in such studies, although they do not provide adequate indication on particle number concentration, especially in the nano–size range. Ultra–fine particle concentration could be assessed by scanning mobility particle counters and condensation nucleus counters. Also, PM measurements could involve active filter sampling with the subsequent analyses of collected deposits for trace metals and organic carbon, yet again, these techniques may not be appropriate considering large movement of equipment and disturbance to the inhabitants.

Based on the short real–time monitoring campaigns, the selected parameters, including background particle concentration, concentration decay rate, indoor–to–outdoor ratio as well as size fraction ratio were found both relevant and optimal for this type of the study. Such combination of several parameters and indicators for the assessment of the effects of the building refurbishment to the indoor air quality proved to be a useful method, which may be recommended to be further applied in the categorization of buildings based on the aerosol concentration in indoor air.

# 5. CONCLUSIONS

1. Particle size distributions and emission factors were determined for 11 powdery building materials widely used in the construction or renovation of buildings. The $PM_{10}$ fraction amounted between 30 and 87 % and the $PM_{2.5}$ portion between 7 and 28 % of the total particulate matter emission. The examined powder being mainly produced by mechanical processes, substantial quantities of fine PM (in the $PM_{2.5}$ range) are emitted into the ambient air during their application processes and may be of potential threat to human health in construction or structural renovation of buildings. The presented emission factors should be utilized for the environmental impact assessment and certification of the powdery building materials.

2. Among tested mechanical operations with cement roofing sheets, crushing appeared to be the most particle emitting operation. Asbestos–cement emitted the lowest amounts of PM (crushing operation, $771.1\pm105.3$ $\#/cm^2/s$). The highest emission of asbestos fibres was generated during crushing operation ($0.059\pm0.011$ $fibre/cm^3$). The emission of cellulose fibres from non–asbestos sheets was 1.8 to 13 times lower in comparison with asbestos fibres release. The highest concentration of asbestos fibres was registered in the $PM_{2.5}$ fraction of aerosol, thus confirming the hazardousness of the asbestos roofing slates. This contradicts to earlier published presumptions on the reduced hazardousness of asbestos fibres due to the binding of fibres to cement matrix.

3. Man-made vitreous fibre emissions from the thermal insulation layer varied in the range of 7–44 $fibre/cm^2/h$ under the airflow through the layer conditions and were dependent on the type of the mineral wool, air flow intensity, and dimensions of microcracks. The fibre penetration was substantial even at minimum areas of micro cracks, resulting in increased levels of MMVFs in settled dust. In residential building MMVF surface concentration ranged 0.11–0.67 $fibre/cm^2$, and began to decrease after two months of completion of construction works.

4. In multifamily buildings, the observed levels of PM concentrations varied greatly among the investigated buildings and apartments, ranging from $4.5\pm5.3$ to $18.1\pm6.4$ $\mu g/m^3$ ($PM_{2.5}$). Mean 24–hr indoor PM concentrations were lower than outdoor concentrations of corresponding locations (average I/O ratio of $PM_{2.5}$ equal to 0.70). Lower outdoor temperature resulted in a more intense fuel burning in the surrounding homes and larger influence of pollution from the transport due to the decreased atmospheric mixing conditions. This, in turn, resulted in the increased penetration of PM into the indoor environment as reflected by higher concentration levels of PM

indoors. The relationship between the effectiveness of natural ventilation and aerosol concentration indoor appeared to be low and statistically insignificant.

5. The before and after refurbishment analysis of aerosol variations in buildings has revealed statistically insignificant differences in PM levels among the investigated 30 apartments. Median $PM_{2.5}/PM_{10}$ ratios (0.65, 0.54) and $PM_{2.5}$ concentration decay rates (0.42, 0.35 $h^{-1}$) were lower after building refurbishment. These changes may be influenced by a more airtight building envelope and decrease of outdoor PM influence. More pronounced differences were registered in extreme cases, where a significant increase in $PM_{2.5}$ concentrations was noticed after refurbishment.

6. A combination of several indicators such as particle decay rate (average of 0.32 $h^{-1}$ for $PM_{2.5}$), I/O ratios (0.7 for $PM_{2.5}$), particle size-selective concentration ratios (average of 0.57 for $PM_{2.5}/PM_{10}$ ratio), and background (nighttime) particle concentrations (5.0 $\mu g/m^3$ for $PM_{2.5}$) was proposed to classify dwelling in multifamily buildings for their IAQ assessment based on the variations in particulate matter concentrations. It may also serve as a tool for the assessment of the effects of building refurbishment on IAQ.

7. The main public concern on the assurance of healthy IAQ in refurbished buildings should be achieved by minimizing the emission of pollutants (avoiding indoor sources of aerosol) and assuring adequate ventilation (the controlled supply of well-filtered outdoor air in case of outdoor pollution events). The assessment of size-segregated indoor aerosol concentrations should be conducted as a supplemental measure in cases where obvious aerosol sources cannot be identified.

## 6. REFERENCES

Abt, E.; Suh, H. H. Relative contribution of outdoor and indoor particle sources to indoor concentrations. *Environmental Science and Technology*. 2000, 34(17), p. 3579-3587.

Afshari, A., *et al.* Emission of phthalates from PVC and other materials. *Indoor Air*. 2004, 14(2), p. 120-128.

Ansari, F. A., *et al.* Monitoring and identification of airborne asbestos in unorganized sectors, India. *Chemosphere*. 2007, 68, p. 716-723.

Barro, R., *et al*. Analysis of industrial contaminants in indoor air: part 1. Volatile organic compounds, carbonyl compounds, polycyclic aromatic hydrocarbons and polychlorinated biphenyls. *Journal of Chromatography*. 2009, 1216(3), p. 540-566.

Baron, P. A. Measurement of airborne fibers: A review. *Industrial Health*. 2001, 39, p. 39-50.

Baxter, L. K., *et al.* Predictors of concentrations of nitrogen dioxide, fine particulate matter, and particle constituents inside of lower socioeconomic status urban homes. *Journal of Exposure Science and Environmental Epidemiology*. 2007, 17, p. 433–444.

Bayram, M.; Bakan, N. D. Environmental exposure to asbestos: from geology to mesothelioma. *Current Opinion in Pulmonary Medicine*. 2014, 20(3), p. 301-307.

Begum, B. A., *et al.* Indoor air pollution from particulate matter emissions in different households in rural areas of Bangladesh. *Building and Environment*. 2009, 44, p. 898–903.

Berge, B. *The ecology of building material. Second edition*. 2009, p. 1-421. ISBN: 978-1-85617-537-1.

Bernstein, J.A., *et al.* The health effects of nonindustrial indoor air pollution. *Journal of Allergy and Clinical Immunology*. 2008, 121, p. 585–591.

BKA. *Housing in Lithuania today*. Ministry of Environment, Lithuania, 2007.

Bone, J., *et al.* From chemical risk assessment to environmental quality management: The Challenge for Soil Protection. *Environmental Science and Technology*. 2011, 45(1), p. 104–110.

Brook, R. D., *et al.* Particulate matter air pollution and cardiovascular disease: an update to the scientific statement from the American Heart Association. *Circulation*. 2010, 121, p. 2331–2378.

Brown, K. W.; Sarnat, J. A.; Koutrakis, P. Concentrations of PM2.5 mass and components in residential and non-residential indoor microenvironments: The sources and composition of particulate exposures study. *Journal of Exposure Science and Environmental Epidemiology*. 2012, 22, p. 161–172.

Brown, S. K. Asbestos exposure during renovation and demolition of asbestos-cement clad buildings. *American Industrial Hygiene Association Journal*. 1987, 48, p. 478-486.

Camilucci, L., *et al.* Man-made mineral fiber exposure in office buildings. *Medicina del Lavoro*. 2001, 92, p. 32-38.

Campopiano, A., *et al.* Risk assessment of the decay of asbestos cement roofs. *The Annals of Occupational Hygiene*. 2009, 53, p. 627–638.

Canpolat, B. R., *et al.* Emission factors of cement industry in Turkey. *Water, Air and Soil Pollution*. 2002, 138, p. 235–252.

Cao, J. J., *et al.* Characterization and source apportionment of atmospheric organic and elemental carbon during fall and winter of 2003 in Xi'an, China. *Atmospheric Chemistry and Physics*. 2005, 5, p. 3127–3137.

Cao, J. J., *et al.* Indoor/Outdoor relationships for organic and elemental carbon in PM2.5 at residential homes in Guangzhou, China. *Aerosol and Air Quality Research*. 2012, 12, p. 902–910.

Chen, C.; Zhao, B. Review of relationship between indoor and outdoor particles: I/O ratio, infiltration factor and penetration factor. *Atmospheric Environment*. 2011, 45, p. 275-288.

Chen, Q.; Hildemann, L. The effects of human activities on exposure to particulate matter and bioaerosols in residential homes. *Environmental Science & Technology*. 2009, 43, p. 4641–4646.

Cheng, Y. H. Comparison of the TSI Model 8520 and Grimm Series 1.108 portable aerosol instruments used to monitor particulate matter in an iron foundry. *Journal of Occupational and Environmental Hygiene*. 2008, 5, p. 157-168.

Commission of the European Communities. *Action plan for energy efficiency: realising the potential*. COM (2006) 545, 2007.

Costello, A., *et al.* Managing the health effects of climate change. *The Lancet*. 2009, 373(9676), p. 1693-1733.

Crump, D.; Dengel, A.; Swainson, M. *Indoor air quality in highly energy efficient homes – a review*. NHBC Foundation, 2009, p. 58, ISBN: 978-1-84806-104-0.

Cullen, R. T., *et al.* Pulmonary and intraperitoneal inflammation induced by cellulose fibres. *Journal of Applied Toxicology*. 2000, 20, p. 49-60.

Cullen, R. T., *et al.* Toxicity of cellulose fibres. *The Annals of Occupational Hygiene*. 2002, 46, p. 81-84.

Deruyterre, A.; Helsen, J.; Baetten, J. The characterization and properties of asbestos cement. *IARC Scientific Publications Series*. 1980, 30, p. 43-51.

Diapouli, E., *et al.* Indoor and outdoor particle number and mass concentrations in Athens. Sources, sinks and variability of aerosol parameters. *Aerosol and Air Quality Research*. 2011, 11, p. 632–642.

Dimitroulopoulou, C. Ventilation in European dwellings: A review. *Building and Environment*. 2012, 47, p. 109-125.

Directive 2002/91/EC of the European Parliament and of the Council of 16 December 2002 on the energy performance of buildings, Official Journal L 001, 04/01/2003 p. 65 - 71.

Directive 2010/31/EU of the European Parliament and of the Council of 19 May 2010 on the energy performance of buildings. Official Journal of the European Union L 153/13, 2010.

Dockery, D. W. Health effects of particulate air pollution. *Annals of Epidemiology*. 2009, 19, p. 257−263.

Dodson, R. F.; Hammar, S. P. *Asbestos: risk assessment, epidemiology, and health effects.* Florida: Boca Raton, 2006. p. 425. ISBN: 978-0849328299.

Dycze, J. Surface of asbestos-cement (AC) roof sheets and assessment of the risk of asbestos release. In *Global Asbestos Congress*, November 19-21, 2004, Tokyo, Japan. 2004, p. 1-9.

Edwards, R. D., *et al.* Personal exposures to VOC in the upper end of the distribution - relationships to indoor, outdoor and workplace concentrations. *Atmospheric Environment*. 2005, 39, p. 2299-2307.

Ehrlich, C., *et al.* PM10, PM2.5 and PM1.0 emissions from industrial plants. Results from measurement programmes in Germany. *Atmospheric Environment*. 2007, 41. p. 6236–6254.

EMEP/CORINAIR Emission Inventory Guidebook – 2007. EEA Technical report No 16/2007. European Environment Agency.

EN 12467:2004. *Fibre-cement flat sheets*. Product specification and test methods. British-Adopted European Standard, 2004, ISBN: 0580450481.

European Agency for Safety and Health at Work. *Expert forecast on emerging chemical risks related to occupational safety and health*. 2009, ISBN 978-92-9191-171-4.

European Commission. *Renovation of buildings using steel technologies*. 2013. ISBN 978-92-79-24950-1.

European Parliament resolution on the European Environment & Health Action Plan 2004-2010, Report, COM (2004) 416, 2005.

Everatt, R. P., *et al.* Occupational asbestos exposure among respiratory cancer patients in Lithuania. *American Journal of Industrial Medicine*. 2007, 50, p. 455-463.

Ferro, A. R.; Kopperud, R. J.; Hildemann, L. M. Elevated personal exposure to particulate matter from human activities in a residence. *Journal of Exposure Analysis and Environmental Epidemiology*. 2004, 14, p. 34–40.

Fittschen, U. E., et *al.* Indoor aerosol determination with respect to a soiling phenomenon in private residences. *Environmental Science & Technology*. 2013, 47, p. 608–615.

Franck, U., *et al.* Respiratory effects of indoor particles in young children are size dependent. *Science of the Total Environment*. 2011, 409, p. 1621–1631.

Frontczak, M.; Andersen, R. V.; Wargocki, P. Questionnaire survey examining factors influencing comfort with indoor environmental quality in Danish housing. *Building and Environment*. 2012, 50, p. 56-64.

Gao, J., *et al.* Volume-based size distribution of accumulation and coarse particles (PM0.1-10) from cooking fume during oil heating. *Building and Environment*. 2013, 59, p. 575-580.

Gaydos, T. M.; Stanier, C.O.; Pandis, S.N. Modelling of in situ ultrafine atmospheric particle formation in the eastern United States. *Journal of Geophysical Research*. 2005, 110, p. 07-12.

Gill, T. E.; Zobeck, T. M.; Stout, J. E. Technologies for laboratory generation of dust from geological materials. *Journal of Hazardous Materials*. 2006, 132. p. 1–13.

Gorner, P., *et al.* Workplace aerosol mass concentration measurement using optical particle counters. *Journal of Environmental Monitoring*. 2012, 14, p. 420–428.

Gualtieri, A. F., *et al.* Ambient monitoring of asbestos in selected Italian living areas. *Journal of Environmental Management*. 2009, 90, p. 3540-3552.

Guarnieri, M.; Balmes, J. R. Outdoor air pollution and asthma. *The Lancet*. 2014, 383(9928), p. 1581–1592.

Gunschera, J., *et al.* Impact of building materials on indoor formaldehyde levels: Effect of ceiling tiles, mineral fiber insulation and gypsum board. *Building and Environment*. 2013, 64, p. 138-145.

Guo, H., *et al.* Characterization of particle number concentrations and PM2.5 in a school: influence of outdoor air pollution on indoor air. *Environmental Science and Pollution Research*. 2010, 17, p. 1268–1278.

Gurskis V. *Statybinės medžiagos.* Lithuanian University of Agriculture, Lithuania: Kaunas. 2008. p. 136. ISBN: 978-9955-896-24-1.

Gustafsson, H. *Building materials identified as major emission sources for indoor air pollutants - a critical review of case studies*, D10, Swedish Council for Building Research, 1992.

Gustafsson, H. *Essential requirements on construction products and current European standardization of emission test methods*. In Conf. Proceedings "Indoor Air Quality and Health for EU Policy", Helsinki, 2007, p. 72-80.

Habre, R., *et al.* Sources of indoor air pollution in New York City residences of asthmatic children. *Journal of Exposure Science and Environmental Epidemiology*. 2014, 24(3), p. 269-278.

Hanninen, O., *et al.* Seasonal patterns of outdoor PM infiltration into indoor environments: review and meta-analysis of available studies from different climatological zones in Europe. *Air Quality, Atmosphere and Health*. 2011, 4, p. 221–233.

Harding, A.H., *et al.* Mortality among British asbestos workers undergoing regular medical examinations (1971–2005). *Occupational and Environmental Medicine*. 2009, 66, p. 487-495.

Harrison, P. T. C., *et al.* Comparative hazards of chrysotile asbestos and its substitutes: A European perspective. *Environmental Health Perspectives*. 1999, 107, p. 607-611.

He, C.; Morawska. L.; Gilbert, D. Particle deposition rates in residential houses. *Atmospheric Environment*. 2005, 39, p. 3891–3899.

He, C.; Morawska, L.; Taplin, L. Particle emission characteristics of office printers. *Environmental Science and Technology*. 2007, 41, p. 6039-6045.

Heinrich, J. Influence of indoor factors in dwellings on the development of childhood asthma. *International Journal of Hygiene and Environmental Health*. 2011, 214, p. 1-25.

Henderson, S. B., *et al.* Differences in lung cancer mortality trends from 1986-2012 by radon risk areas in British Columbia, Canada. *Health Physics*. 2014, 106(5), p. 608-613

Ho, K. F., *et al.* Indoor/outdoor relationships of organic carbon (OC) and elemental carbon (EC) in PM2.5 in roadside environment of Hong Kong. *Atmospheric Environment*. 2004, 38, p. 6327–6335.

Hodgson, J. T., *et al.* The expected burden of mesothelioma mortality in Great Britain from 2002–2050. *British Journal of Cancer*. 2005, 92, p. 583-593.

Huang, H., *et al.* Spatial variation and relationship of indoor/outdoor PM2.5 at residential homes in Guangzhou city, China. *Aerosol and Air Quality Research*. 2007, 7, p. 518–530.

Hussein, T., *et al.* Indoor and outdoor particle size characterization at a family house in Espoo-Finland. *Atmospheric Environment*. 2005, 39(20), p. 3697-3709.

Hussein, T., *et al.* Particle size characterization and emission rates during indoor activities in a house. *Atmospheric Environment*. 2006, 40, p. 4285–4307.

International Agency for Research on Cancer (IARC). *Monographs on the Evaluation of Carcinogenic Risks to Humans, Man–Made Mineral Fibres*, Volume 81, 2002.

ISO 23210:2009. *Stationary source emissions -- Determination of PM10/PM2.5 mass concentration in flue gas - Measurement at low concentrations by use of impactors*. 2009, p. 42.

Jantunen, M.; Oliveira Fernandes, E.; Carrer, P.; Kephalopoulos, S. *Promoting actions for healthy indoor air (IAIAQ). European Commission Directorate General for Health and Consumers*, Luxembourg, 2011.

Jantunen, M.; Carrer, P. *Introduction to the EnVIE concept and overview of EnVIE programme*. In Proceedings of the first EnVIE conference on indoor air quality and health for EU policy, 2007, p. 41-52.

Jensen, A.; Knudsen, H. *Total health assessment of chemicals in indoor climate from various consumer products*. Survey of chemical substances in consumer products nº. 75, Danish Ministry of the Environment, 2006.

Janssens, A.; Hens, H. Interstitial condensation due to air leakage: a sensitivity analysis. *Journal of Thermal Envelope and Building Science*. 2003, 27(1), p. 15–29.

Jung, K. H., *et al.* Effects of heating season on residential indoor and outdoor polycyclic aromatic hydrocarbons, black carbon, and particulate matter in an urban birth cohort. *Atmospheric Environment*. 2010, 44, p. 4545–4552.

Kakooei, H., *et al.* Assessment of airborne asbestos fiber concentrations in urban area of Tehran, Iran. *Air Quality, Atmosphere and Health*. 2009, 2, p. 39-45.

Kaya, E.; Hogg, R.; Mutmansky, J. M. Evaluation of procedures for production of dust samples for biomedical research. *Applied Occupational and Environmental Hygiene*. 1996, 11, p. 745–750.

Kearney, J., *et al.* Residential indoor and outdoor ultrafine particles in Windsor, Ontario. *Atmospheric Environment*. 2011, 45, p. 7583–7593.

Klepeis, N. E., *et al.* The National Human Activity Pattern Survey (NHAPS): A resource for assessing exposure to environmental pollutants. *Journal of Exposure Analysis and Environmental Epidemiology*. 2001, 11(3), p. 231-252.

Klepeis, N. E., *et al.* Determining size-specific emission factors for environmental tobacco smoke particles. *Aerosol Science and Technology*. 2003, 37, p. 780–79.

Kim, S., *et al.* TVOC and formaldehyde emission behaviours from flooring materials bonded with environmental-friendly MF/PVAc hybrid resins. *Indoor Air*. 2007, 17(5), p. 404-415.

Kliucininkas, L., *et al.* Indoor and outdoor concentrations of fine particles, particle-bound PAHs and volatile organic compounds in Kaunas, Lithuania. *Journal of Environmental Monitoring*. 2011, 13, p. 182-191.

Koistinen, K. J., *et al.* Sources of fine particulate matter in personal exposures and residential indoor, residential outdoor and workplace microenvironments in the Helsinki phase of the EXPOLIS study. *Scandinavian Journal of Work, Environment & Health*. 2004, 30, p. 36–46.

Kovanen, K., *et al.* Man made mineral fiber emission from HVAC-components. Proceedings of Clima 2007 WellBeing Indoors, 10-14 June Helsinki, Finland. 2007, p. 7.

Krakowiak, E., *et al.* Environmental exposure to airborne asbestos fibres in a highly urbanized city. *Annals of Agricultural and Environmental Medicine*. 2009, 16, p. 121-128.

La Vecchia, C.; Boffetta, P. Role of stopping exposure and recent exposure to asbestos in the risk of mesothelioma. *European Journal of Cancer Prevention*. 2012, 21(3), p. 227-230.

Laden, F., *et al.* Reduction in fine particulate air pollution and mortality-extended follow-up of the Harvard six cities study. *American Journal of Respiratory and Critical Care Medicine*. 2006, 173, p. 667-672.

Lai, H. K., *et al.* Personal exposures and microenvironment concentrations of PM2.5, VOC, NO2 and CO in Oxford, UK. *Atmospheric Environment*. 2004, 38, p. 6399–6410.

Lai, S. C., *et al.* Characteristics of residential indoor carbonaceous aerosols: a case study in Guangzhou, Pearl river delta region. *Aerosol and Air Quality Research*. 2010, 10, p. 472–478.

Lamonaca, F. *et al.* Monitoring of indoor radon pollution. *Measurement.* 2014, 47, p. 228-233.

Layton, D. W.; Beamer, P. I. Migration of contaminated soil and airborne particulates to indoor dust. *Environmental Science and Technology*. 2009, 43, p. 8199-8205.

Lee, S. H.; Li, W. M.; Ao, C. H. Investigation of indoor air quality at residential homes in Hong Kong-case study. *Atmospheric Environment*. 2002, 36, p. 225–237.

Leech, J. A., *et al.* It's about time: a comparison of Canadian and American time-activity patterns. *Journal of Exposure Analysis and Environmental Epidemiology*. 2002, 12(6), p. 427-432.

Lianou, M., *et al.* Temporal variations of atmospheric aerosol in four European urban areas. *Environmental Science and Pollution Research*. 2011, 18, p. 1202–1212.

Lim, S. S., *et al.* A comparative risk assessment of burden of disease and injury attributable to 67 risk factors and risk factor clusters in 21 regions, 1990–2010: a systematic analysis for the Global Burden of Disease Study 2010. *The Lancet*. 2012, 380, p. 2224–2260.

Lippman, M. Effects of fiber characteristics on lung deposition, retention, and disease. *Environmental Health Perspectives*. 1990, 88, 311-317.

Liu, D.; Nazaroff, W. W. Particle penetration through building cracks. *Aerosol Science and Technology*. 2003, 37, p. 565-573.

Liu, D. L.; Nazaroff, W. W. Modelling pollutant penetration across building envelopes. *Atmospheric Environment*. 2001, 35(26), p. 4451-4462.

Lonati, G., *et al.* Pedestrian exposure to size-resolved particles in Milan. *Journal of the Air & Waste Management Association*. 2011, 61, p. 1273–1280.

Long, C. M., *et al.* Using time- and size-resolved particulate data to quantify indoor penetration and deposition behavior. *Environmental Science & Technology*. 2001, 35, p. 2089–2099.

Lopez, E.; Schuhmacher, M.; Domingo, J. L. Human health risks of petroleum-contaminated groundwater. *Environmental Science and Pollution Research*. 2008, 15, p. 278-288.

LR aplinkos ministro ir LR sveikatos apsaugos ministro 2001 m įsakymo nr. 591/640 *„Dėl aplinkos oro užterštumo normų nustatymo" pakeitimo*, 2010, Vilnius, Lietuva. Valstybės žinios, 2001-12-19, Nr. 106-3827.

LST EN 15051:2006. *Workplace exposure – Measurement of the dustiness of bulk material – Part 3: Continuous drop method*, 2006.

Mann, H. B.; Whitney, D. R. On a test of whether one of two random variables is stochastically larger than the other. The Annals of Mathematical Statistics, 1947, 18, p. 50–60.

Martuzevicius, D., et al. Traffic-related PM2.5 aerosol in residential houses located near major highways: indoor versus outdoor concentrations. *Atmospheric Environment*. 2008, 42, p. 6575–6585.

Massey, D., *et al.* Indoor/outdoor relationship of fine particles less than 2.5 μm (PM2.5) in residential homes locations in central Indian region. *Building and Environment*. 2009, 44, p. 2037–2045.

Massey, D., *et al.* Seasonal trends of PM10, PM5.0, PM2.5 & PM1.0 in indoor and outdoor environments of residential homes located in North-Central India. *Building and Environment*. 2012, 47, p. 223–231.

Mendell, M. J., *et al.* Indicators of moisture and ventilation system contamination in U.S. office buildings as risk factors for respiratory and mucous membrane symptoms: analyses of the EPA BASE data. *Journal of Occupational and Environmental Hygiene*. 2006, 3(5), p. 225-233.

Meng, Q. Y., *et al.* Influence of ambient (outdoor) sources on residential indoor and personal PM2.5 concentrations: Analyses of RIOPA data. *Journal of Exposure Analysis and Environmental Epidemiology*. 2005, 15, p. 17–28.

Milner, J., *et al.* Modelling inhalation exposure to combustion-related air pollutants in residential buildings: Application to health impact assessment. *Environment International*. 2011, 37, p. 268–279.

Morawska, L., *et al.* Relation between particle mass and number for submicrometer airborne particles. *Atmospheric Environment*. 1999, 33, p. 1983-1990.

Morawska, L., *et al.* The relationship between indoor and outdoor airborne particles in the residential environment. *Atmospheric Environment*. 2001, 35, p. 3463–3473.

Morawska, L.; Salthammer, T. *Indoor environment. Airborne particles and settled dust.* WILEY-VCH Verlag GmbH, 2003, p. 450. ISBN: 978-3-527-30525-4.

Morawska, L., *et al.* Ambient nano and ultrafine particles from motor vehicle emissions: characteristics, ambient processing and implications on human exposure. *Atmospheric Environment*. 2008, 42(35), 8113-8138.

Mowat, F.; Weidling, R.; Sheehan, P. Simulation tests to assess occupational exposure to airborne asbestos from asphalt-based roofing products. *The Annals of Occupational Hygiene*. 2007, 51, p. 451-462.

Muhle, H.; Ernst, H.; Bellmann, B. Investigation of the durability of cellulose fibres in rat lungs. *The Annals of Occupational Hygiene*. 1997, 41, p. 184–188.

Muleski, G. E.; Cowherd, C. Jr.; Kinsey, J. S. Particulate emissions from construction activities. *Journal of the Air and Waste Management Association*. 2005, 55, p. 772-783.

Nazaroff, W. W. Indoor particle dynamics. *Indoor Air*. 2004, 14, p. 175–183.

Nazaroff, W. W.; Weschler, C. Cleaning products and air fresheners: exposure to primary and secondary air pollutants. *Atmospheric Environment*. 2004, 38, p. 2841-2865.

Nicole, W. Cooking Up Indoor Air pollution emissions from natural gas stoves. *Environmental Health Perspectives*. 2014, 122(1), p. A27.

Oliveira Fernandes, E.; Gustafsson, H.; Seppanen, O.; Crump, D.; Silva, G. V. *Co-ordination action on indoor air quality and health effects. WP3 Technical Report: Characterisation of spaces and sources*. 2008. Project no. SSPE-CT-2004-502671.

Oreszczyn, T.; Mumovic, D.; Ridley, I.; Davies, M. The reduction in air infiltration due to window replacement in UK dwellings: Results of a field study and telephone survey. *International Journal of Ventilation*. 2005, 4(1), p. 71-78.

Owen, M. K.; Ensor, D. S.; Sparks, L. E. Airborne particle sizes and sources found in indoor air. *Atmospheric Environment*. 1992, 26A, p. 2149–2162.

Ozkaynak, H., *et al.* Personal exposure to airborne particles and metals: Results from the particle team study in Riverside, California. *Journal of Exposure Analysis and Environmental Epidemiology*. 1996, 6, p. 57–78.

Pagels, J., *et al.* Chemical composition and mass emission factors of candle smoke particles. *Journal of Aerosol Science*. 2009, 40, p. 193–208.

Palomaki, E., *et al.* Decreasing irritation symptoms by replacing partially coated acoustic glass wool boards with fully coated boards. *Scandinavian Journal of Work, Environment & Health Supplements*. 2008, 4, p. 64-68.

Pastuszka, J. S. Emission of airborne fibers from mechanically impacted asbestos-cement sheets and concentration of fibrous aerosol in the home environment in Upper Silesia, Poland. *Journal of Hazardous Materials*. 2009, 162, p. 1171-1177.

Petry, T., *et al.* Human health risk evaluation of selected VOC, SVOC and particulate emissions from scented candles. *Regulatory Toxicology and Pharmacology*. 2014, 69(1), p. 55-70.

Pilkyte, L.; Butkus, D.; Morkunas, G. Assessment of external dose indoors in Lithuania. *Radiation Protection Dosimetry*. 2006, 121(2), p. 140-147.

Pitz, M., *et al.* Variability of apparent particle density of an urban aerosol. *Environmental Science & Technology*. 2003, 37, p. 4336-4342.

Pope, C. A. Review: Epidemiological basis for particulate air pollution health standards. *Aerosol Science and Technology*. 2000, 32, p. 4–14.

Pope, C. A.; Dockery, D. W. Health effects of fine particulate air pollution: lines that connect. *Journal of the Air and Waste Management Association*. 2006, 56, p. 709−742.

Poupard, O., *et al.* Statistical analysis of parameters influencing the relationship between outdoor and indoor air quality in schools. *Atmospheric Environment*. 2005, 39, p. 2071–2080.

Preat, B. *Suveillance de Chantiers avec fibres-ciment.* Instituut voor Reddingswezen, Ergonomie en Arbeidshygiene, Hasselt, Belgium, Report Ref. 2240-F, 1993.

Puustinen, A., *et al.* Spatial variation of particle number and mass over four European cities. *Atmospheric Environment*. 2007, 41, p. 6622–6636.

Raja, S., *et al.* Resuspension of indoor aeroallergens and relationship to lung inflammation in asthmatic children. *Environment International*. 2010, 36, p. 8-14.

Ridley, I.; Fox, J.; Oreszczyn, T.; Hong, S. The Impact of Replacement Windows on Air Infiltration and Indoor Air Quality in Dwellings. *International Journal of Ventilation*. 2003, 1(3), p. 209-218.

Ruckerl, R., *et al.* Health effects of particulate air pollution: A review of epidemiological evidence. *Inhalation Toxicology*. 2011, 23, p. 555–592.

Ryan, T. J.; Beaucham, C. Dominant microbial volatile organic compounds in 23 US homes. *Chemosphere*. 2013, 90(3), p. 977-985.

Rydock, J. P.; Skaret, E. Case study of sub-slab depressurization for a building located over VOC-contaminated ground. *Building and Environment*. 2002, 37, p. 1343-1347.

Salonen, H. J., *et al.* Man-made vitreous fibers in office buildings in the Helsinki area. *Journal of Occupational and Environmental Hygiene*. 2009, 6, p. 624–631.

Santacatalina, M., *et al.* Impact of fugitive emissions in ambient PM levels and composition: A case study in Southeast Spain. *Science of the Total Environment*. 2010, 408. p. 4999-5009.

Sawant, A. A., *et al.* Characterization of PM2.5 and selected gas-phase compounds at multiple indoor and outdoor sites in Mira Loma, California. *Atmospheric Environment*. 2004, 38, p. 6269–6278.

Schneider, T. *Synthetic vitreous fibers (SVF's).* New York, McGraw-Hill, 2000.

Shapiro, S. S.; Wilk, M. B. An analysis of variance test for normality (complete samples). *Biometrika*. 1965, 52, p. 591-611.

Shehabi, A., *et al.* Particle concentrations in data centers. *Atmospheric Environment*. 2008, 42, p. 5978–5990.

Shen, R.; Lin, Z.; Zhang, Y. *Development of non-asbestos fibre cement products in China.* In 10th International Inorganic-Bonded Fiber Composites Conference, Sao Paulo, Brazil, November 15-18, 2006, p. 68-78.

Shimada, Y.; Matsuoka, Y. Analysis of indoor PM2.5 exposure in Asian countries using time use survey. *Science of the Total Environment*. 2011, 409. p. 5243–5252.

Shneider, T., *et al.* Dust in buildings with man – made mineral fiber ceiling boards. *Scandinavian Journal of Work, Environment & Health*. 1990, 16(6), p. 434-439.

Smolianskiene, G.; Adamoniene, D.; Seskauskas, V. Studies on occupational asbestos in Lithuania: Achievements and problems. *Indoor and Built Environment*. 2005, 14, p. 331-335.

Singer, B.C., *et al.* Cleaning products and air fresheners: emissions and resulting concentrations of glycol ethers and terpenoids. *Indoor Air*. 2006, 16, p. 179–191.

Spengler, J. D., *et al.* Climate change, the indoor environment, and health. Report Brief, National Academy of Sciences, 2011.

Spilak, M. P., *et al.* Evaluation of building characteristics in 27 dwellings in Denmark and the effect of using particle filtration units on PM2.5 concentrations. *Building and Environment*. 2014, 73, p. 55-63.

Spruyt, M., *et al.* Influence of air fresheners on the indoor air quality. Research Unit Environmental Measurement, report 2006/MIM/R/032. Belgian Federal Public Service of Health, Food Chain safety and Environment, 2006.

Spurny, K. R. On the emission of fibrous particles from weathered and corroded asbestos-cement products. *Environmental Research*. 1989, 48, p. 110-116.

SQUARE – *A System for Quality Assurance when Retrofitting Existing Buildings to Energy Efficient Buildings*. 2010. Energy improvement measures and their effect on the indoor environment. 2010 p. 47.

Stankevicius, V.; Karbauskaite, J.; Monstvilas, E. The development of reference values for energy certification of buildings in Lithuania. *Energy and Buildings*. 2007, 39, p. 284-288.

Stranger, M.; Potgieter-Vermaak, S. S.; Van Grieken, R. Particulate matter and gaseous pollutants in residences in Antwerp, Belgium. *Science of the Total Environment*. 2009, 407, p. 1182–1192.

Stymne, H.; Emenius, G.; Boman, C. A. Passive tracer gas measurement of the long term variation of ventilation in three Swedish dwellings. *International Journal of Ventilation*. 2006, 5(3), p. 333–344.

Thatcher, T., *et al.* A concentration rebound method for measuring particle penetration and deposition in the indoor environment. *Aerosol Science and Technology*. 2003, 37, p. 847-864.

Tippayawong, N., *et al.* Indoor/outdoor relationships of size-resolved particle concentrations in naturally ventilated school environments. *Building and Environment*. 2009, 44, p. 188–197.

Tittarelli, A., *et al.* Estimation of particle mass concentration in ambient air using a particle counter. *Atmospheric Environment*. 2008, 42, p. 8543–8548.

Tossavainen, A. Asbestos, asbestosis and cancer: the Helsinki criteria for diagnosis and attribution. Consensus report. *Scandinavian Journal of Work, Environment & Health*. 1997, 23, p. 311-316.

Uhde, E.; Salthammer, T. Impact of reaction products from building materials and furnishings on indoor air quality-A review of recent advances in indoor chemistry. *Atmospheric Environment*. 2007, 41, p. 3111–3128.

Valarino, J., *et al.* Quantifying synthetic vitreous fiber surface contamination in office Buildings. *American Industrial Hygiene Association Journal*. 2003, 64(1), p. 80–87.

Vitez, T.; Travnicek, P. Particle size distribution of sawdust and wood shaving mixture. *Research in Agricultural Engineering*. 2010, 56(4), p. 154-158.

Vuyst, P. De., *et al.* Respiratory health effects of man–made vitreous (mineral) fibres. *European Respiratory Journal*. 1995, 8, p. 2149–2173.

Wallace, L., *et al.* Continuous weeklong measurements of personal exposures and indoor concentrations of fine particles for 37 health-impaired North Carolina residents for up to four seasons. *Atmospheric Environment*. 2006, 40, p. 399–414.

Wang, L.; Morawska, L. *Analysis of particle number concentration in a mechanically ventilated office equipped with a printer*. In Proceedings of Indoor Air 2008, Copenhagen, Denmark, paper ID: 196, 2008.

Wargocki, P.; Wyon, D. P.; Fanger, P.O. The performance and subjective responses of call-center operators with new and used supply air filters at two outdoor air supply rates. *Indoor Air*. 2004, 14(8), p. 7-16.

Warheit, D. B., *et al.* Two-week inhalation study in rats with cellulose fibers. In: Advances in the prevention of occupational respiratory disease, eds. by Chiyotani K, Hosoda Y, Aizawa Y, 1998, p. 579–582.

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Warheit, D. B.; Reed, K. L.; Webb, T. R. Man-made respirable-sized organic fibers: What do we know about their toxicological profiles? *Industrial Health*. 2001, 39, p. 119-125.

Weber, K., *et al.* Airborne in-situ investigations of the Eyjafjallajökull volcanic ash plume on Iceland and over north-western Germany with light aircrafts and optical particle counters. *Atmospheric Environment*. 2012, 48, p. 9-21.

Weichenthal, S.; Dufresne, A.; Infante-Rivard, C. Indoor ultrafine particles and childhood asthma: exploring a potential public health concern. *Indoor Air*. 2007, 17, p. 81–91.

Weschler, C. *Ozone's impact on public health: contributions from indoor exposures to ozone and products of ozone-initiated chemistry.* The National Institute of Environmental Health Sciences. National Institute of Health U.S. Department of Health and Human Services, 2006, 41 p.

Wheeler, A. J., *et al.* Personal, indoor, and outdoor concentrations of fine and ultrafine particles using continuous monitors in multiple residences. *Aerosol Science and Technology*. 2011, 45, p. 1078–1089.

Wheeler, A. J., *et al.* Residential indoor and outdoor coarse particles and associated endotoxin exposures. *Atmospheric Environment*. 2011, 45, p. 7064–7071.

Wichmann, J., *et al.* PM2.5, soot and NO2 indoor-outdoor relationships at homes, pre-schools and schools in Stockholm, Sweden. *Atmospheric Environment*. 2010, 44, p. 4536–4544.

Wilcoxon, F. Individual comparisons by ranking methods. *Biometric Bulletin*. 1945, 1, 80–83.

World Health Organisation (WHO). *Determination of airborne fibre number concentration. A recommended method by phase-contrast optical microscopy (membrane filter method).* WHO, Geneva, 1997.

World Health Organisation (WHO). *Air quality guidelines for Europe. Geneva. Second edition*. 2000, ISBN 9289010827.

World Health Organisation (WHO). *Silicosis.* Fact sheet No. 238, 2000.

World Health Organisation (WHO). *Environmental burden of disease associated with inadequate housing. A method guide to the quantification of health effects of selected housing risks in the WHO European Region*. 2011, ISBN: 9789289002394.

World Health Organisation (WHO) Regional Office for Europe. Fourth Ministerial Conference on Environment and Health, Budapest, Hungary, 23-25 June, Final Conference Report, 2004.

World Health Organisation (WHO) Regional Office for Europe. WHO Symposium on Housing and Health in Europe. Bonn, Germany. Jun 6-8. [Report], Symposium, Bonn, Germany, 2001.

World Health Organisation (WHO). *Air quality guidelines for particulate matter, ozone, nitrogen dioxide and sulphur dioxide*. Global update, 2005.

World Health Organisation (WHO) Regional Office for Europe. Preliminary results of the WHO Frankfurt housing intervention project. 2008.

Wright, L. S.; Phipatanakul, W. Environmental remediation in the treatment of allergy and asthma: latest updates. *Current Allergy and Asthma Reports*. 2014, 14(3), p. 1-10.

Yu, B. F., *et al.* Review of research on air-conditioning systems and indoor air quality control for human health. *International Journal of Refrigeration*. 2009, 32(1), p. 3-20.

Zhang, J.; Smith, K. R. Indoor air pollution: a global health concern. *British Medical Bulletin*. 2003, 68, p. 209–225.

Ziegler-Skylakakis, K. Approaches for the development of occupational exposure limits for man-made mineral fibres (MMMFs). *Mutation Research*. 2004, 41, p. 553-537.

Zwozdziak, A., *et al.* Infiltration or indoor sources as determinants of the elemental composition of particulate matter inside a school in wroc1aw, Poland? *Building and Environment*. 2013, 66(0), p. 173-180.

**Internet access:**

Bridle, J.; Stone, S. *Casitile, the new asbestos: Time to clear the air and save £20 billion.* Internet article on the asbestos watchdog internet site. 2006. [interactive] [previewed 2011 03 12]. Web access:

http://www.chrysotile.com/data/Casitile,%20The%20New%20Asbestos%20Revised.pdf

Burdett G. *Investigation of the chrysotile fibres in an asbestos cement sample*. Derbyshire: Harpur Hill; 2006. [interactive] [previewed 2011 05 11]. Web access: http://www.hse.gov.uk/research/hsl_pdf/2007/hsl0711.pdf.

Gray, J. *Sustainable build, pollution from construction*, [interactive] [previewed 2011 11 09]. Web access:

http://www.sustainablebuild.co.uk/PollutionFromConstruction.html

## LIST OF PUBLICATIONS ON THE TOPIC OF THE DISSERTATION

### Publications corresponding to the list of the Institute of Science Information (ISI) database

1. **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: 2,430, AIF: 2,073 (E, 2012)].

2. **Prasauskas, Tadas;** Martuzevičius, Dainius; Krugly, Edvinas; Kliučininkas, Linas; Kireitseu, Maksim; Zerrath, Axel. Comparative characterization of particle emissions from asbestos and non-asbestos cement roof slates // Building and Environment. Oxford: Pergamon-Elsevier Science Ltd. ISSN 0360-1323. 2011, Vol. 46, iss. 11, p. 2295-2302. [Science Citation Index Expanded (Web of Science)]. [0,236]. [IF (E): 2,131 (2010)].

### Publications referred in International databases

1. **Prasauskas, Tadas;** Zemaityte, Aida; Krugly, Edvinas; Ciuzas, Darius; Martuzevicius, Dainius. Characterization of particle size distributions of powdery building material aerosol generated by fluidization and gravitation // Environmental research, engineering and management = Aplinkos tyrimai, inžinerija ir vadyba. Kaunas : KTU. ISSN 1392-1649. 2012, Nr. 3(61), p. 50-57.

### List of the presentations in the international conferences

1. **Prasauskas, Tadas;** Krugly, Edvinas; Ciuzas, Darius; Sidaraviciute, Ruta; Zaluda, Darius; Jurelionis, Andrius; Seduikyte, Lina; Gagyte, Laura; Martuzevicius, Dainius. (2014). Characterization of Mineral Fibre Emissions from Processes of Building Refurbishment and Operation // World Sustainable Buildings 2014, Barcelona, October 28-30, Spain.

2. **Prasauskas, Tadas;** Martuzevicius, Dainius; Krugly, Edvinas; Ciuzas, Darius; Sidaraviciute, Ruta; Stankevicius, Kazimieras; Haverinen-Shaughnessy, Ulla. Characterization of multifamily buildings by representative indicators of indoor PM // 3rd Workplace and Indoor Aerosols Conference: Institute of Immunology and Experimental Therapy, 13-16 May 2014: proceedings, 2014. p. 65. [M.kr. 04T].
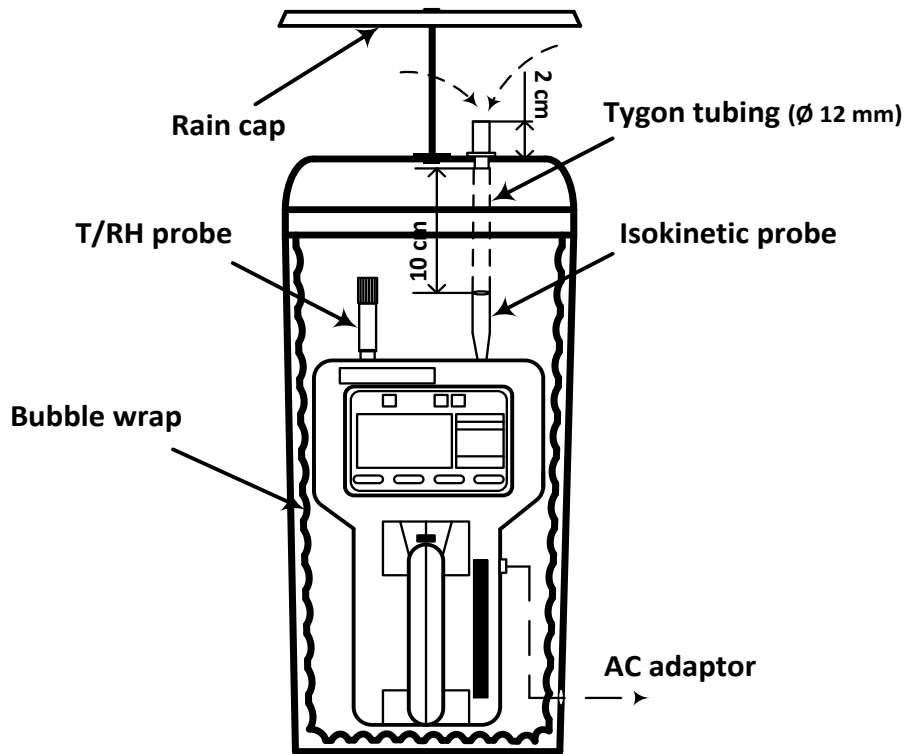
3. **Prasauskas, Tadas;** Ciuzas, Darius; Stankevicius, K; Krugly, Edvinas; Martuzevicius, Dainius. Comparison of particulate matter concentrations before and after refurbishment in multifamily buildings // Chemistry and Chemical Technology 2014 proceedings: Environmental chemistry and technology, 25 April, 2014, Kaunas: Kaunas University of Technology, Lithuania. ISSN 2351-5643. p. 38-41. [M.kr. 04T].

4. Martuzevičius, Dainius; **Prasauskas, Tadas;** Turunen, Mari; Leivo, Virpi; Aaltonen, Anu; Krugly, Edvinas; Tichonovas, Martynas; Sidaravičiūtė, Rūta; Čiužas, Darius; Pekkonen, Maria; Kiviste, Mihkel; Pilkytė, Laima; Ladygienė, Rima; Haverinen-Shaughnessy, Ulla. Improving Energy Efficiency of Housing Stock: Demonstration of Impacts on Indoor Environmental Quality and Public Health in Northern Europe // ASHRAE IAQ 2013 proceedings: Environmental health in low-energy buildings, October 15-18, 2013, Vancouver, British Columbia, Canada. Atlanta, GA: ASHRAE. ISSN 2166-4870. 2013, p. 561-565.

5. Martuzevičius, Dainius; **Prasauskas, Tadas;** Krugly, Edvinas; Čiužas, Darius; Turunen, Mari; Leivo, Virpi; Aaltonen, Anu; Haverinen-Shaughnessy, Ulla. Background indoor/outdoor PM levels in Finnish and Lithuanian multifamily buildings // Workplace and indoor aerosols conference: Lund University, 19-20 April 2012: proceedings, 2011. p. 39. [M.kr. 04T].

6. Martuzevičius, Dainius; **Prasauskas, Tadas;** Turunen, Mari; Krugly, Edvinas; Šeduikytė, Lina; Täubel, M.; Leivo, Virpi; Haverinen-Shaughnessy, Ulla. Assessment of PM levels in the process of energy efficiency improvement of apartment buildings: measurement approach in INSULAtE project // NOSA & FAAR 2011: Nordic Aerosol Symposium, November 9-11 2011, Tampere, Finland: programme, conference proceedings. Tampere: Tampere University of Technology, 2011. p. 59. [M.kr. 04T].

7. Leivo, Virpi; Turunen, Mari; **Prasauskas, Tadas;** Pekkonen, Maria; Martuzevičius, Dainius; Aaltonen, Anu; Haverinen-Shaughnessy, Ulla. Assessment of improving energy efficiency of buildings: impacts on indoor environmental quality and public health in Europe // SB 11 Helsinki: World Sustainable Building Conference, 18-21 October, 2011: proceedings, Vol. 2. Helsinki: Finnish Association of Civil Engineers RIL, 2011. ISBN 9789517585316. p. 324-332. [M.kr. 04T].

8. **Prasauskas, Tadas;** Martuzevičius, Dainius; Krugly, Edvinas; Kliučininkas, Linas. Characterization of particle size distributions of powdery building materials aerosol // EAC 2011: European aerosol conference handbook, 4-9 September, 2011, Manchester, England. Manchester: Aerosol Society, 2011. p. 734. [M.kr. 04T].

9.  **Prasauskas, Tadas;** Krugly, Edvinas; Martuzevičius, Dainius; Kliučininkas, Linas. Particle size distribution and asbestos fibres emission of aerosol from treatment process of asbestos-cement sheets // IAC2010: International Aerosol Conference, August 29 - September 3, Helsinki, Finland: abstracts. Helsinki: Finnish Association for Aerosol Research, 2010. p. 1. [M.kr. 04T].

**LIST OF ANNEXES**

**ANNEX 1.** Cross–section of the cooler box used as an enclose compartment for the PM measurement
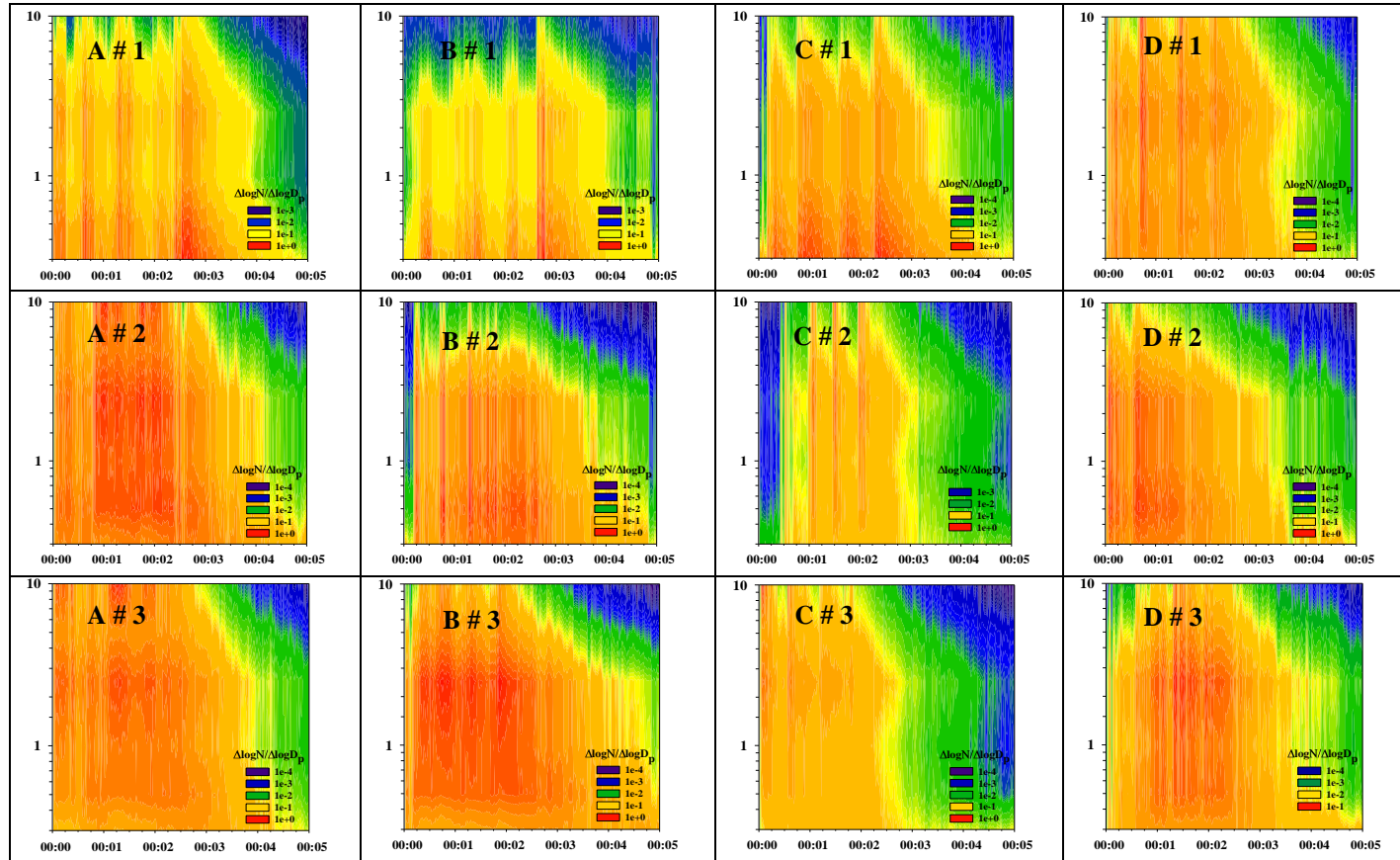
**ANNEX 2.** Photos of PM *in situ* measurements

**ANNEX 3.** The external modification of multifamily building before and after refurbishment

**ANNEX 4.** Distribution of PM number concentration in time for all mechanical operations of different types of sheets (A – crushing; B – rubbing C – rasping; D – scrubbing)

**ANNEX 5.** Temporal variation of indoor and outdoor PM concentration, based on the coefficient of variation (CV)

| | Indoor | | | Outdoor | | |
|---|---|---|---|---|---|---|
| | $PM_1$ | $PM_{2.5}$ | $PM_{10}$ | $PM_1$ | $PM_{2.5}$ | $PM_{10}$ |
| B1A1 | 0.28 | 0.36 | 0.64 | 0.22 | 0.24 | 0.31 |
| B1A2 | 0.23 | 0.29 | 0.82 | 0.23 | 0.25 | 0.23 |
| B1A3 | 0.31 | 0.33 | 0.84 | 0.25 | 0.26 | 0.30 |
| B1A4 | 0.54 | 0.49 | 0.54 | - | - | - |
| B1A5 | 0.29 | 0.34 | 0.55 | 0.24 | 0.25 | 0.26 |
| B2A1 | 0.64 | 0.67 | 0.75 | - | - | - |
| B2A2 | 0.09 | 0.18 | 0.93 | 0.22 | 0.24 | 0.31 |
| B2A3 | 0.17 | 0.22 | 0.53 | 0.22 | 0.24 | 0.31 |
| B2A4 | 0.65 | 0.76 | 1.06 | 0.62 | 0.42 | 0.40 |
| B2A5 | 0.48 | 0.34 | 0.59 | 0.78 | 0.51 | 0.37 |
| B2A6 | 0.60 | 0.49 | 1.87 | 0.70 | 0.47 | 0.42 |
| B3A1 | 0.53 | 0.84 | 1.19 | 0.45 | 0.48 | 0.49 |
| B3A2 | 1.36 | 1.33 | 1.06 | 0.63 | 0.63 | 0.78 |
| B3A3 | 0.29 | 0.28 | 0.25 | 0.27 | 0.28 | 0.26 |
| B3A4 | 0.17 | 0.18 | 0.34 | 0.23 | 0.25 | 0.21 |
| B4A1 | 0.30 | 0.27 | 0.25 | 0.17 | 0.19 | 0.21 |
| B4A2 | 0.23 | 0.24 | 0.30 | 0.23 | 0.30 | 0.35 |
| B4A3 | 0.54 | 0.47 | 0.40 | 0.21 | 0.28 | 0.34 |
| B4A4 | 0.25 | 0.26 | 0.33 | 0.27 | 0.30 | 0.31 |
| B4A5 | 0.56 | 0.58 | 0.84 | 0.28 | 0.31 | 0.30 |
| B5A1 | 1.55 | 1.36 | 1.06 | 0.71 | 0.67 | 0.77 |
| B5A2 | 0.54 | 0.79 | 1.50 | 0.51 | 0.43 | 0.40 |
| B5A3 | 0.45 | 0.48 | 0.87 | - | - | - |
| B5A4 | 0.22 | 0.21 | 0.61 | 0.49 | 0.46 | 0.43 |
| B5A5 | 0.48 | 0.49 | 0.59 | - | - | - |
| B6A1 | 0.47 | 0.45 | 0.71 | 0.64 | 0.57 | 0.47 |
| B6A2 | 0.50 | 0.49 | 0.95 | 0.66 | 0.58 | 0.53 |
| B6A3 | 0.90 | 0.77 | 0.67 | - | - | - |
| B6A4 | 0.44 | 0.44 | 0.42 | 0.61 | 0.68 | 0.66 |
| B6A5 | 0.44 | 0.44 | 0.53 | 0.57 | 0.62 | 1.23 |
| B7A1 | 0.27 | 0.32 | 0.67 | 0.33 | 0.33 | 0.26 |
| B7A2 | 0.79 | 0.75 | 0.75 | 0.64 | 0.60 | 0.44 |
| B7A3 | 0.93 | 1.37 | 1.43 | 0.66 | 0.62 | 0.50 |

| | Indoor | | | Outdoor | | |
|---|---|---|---|---|---|---|
| | $PM_1$ | $PM_{2.5}$ | $PM_{10}$ | $PM_1$ | $PM_{2.5}$ | $PM_{10}$ |
| **B7A4** | 0.26 | 0.26 | 0.33 | 0.55 | 0.60 | 0.45 |
| **B7A5** | 0.48 | 0.48 | 0.72 | 0.53 | 0.58 | 0.43 |
| **B8A1** | 1.26 | 1.21 | 1.70 | 0.39 | 0.39 | 0.47 |
| **B8A2** | 1.07 | 1.24 | 1.43 | 0.54 | 1.43 | 2.19 |
| **B8A3** | 0.70 | 0.79 | 1.07 | 0.40 | 0.49 | 0.78 |
| **B8A4** | 0.80 | 1.49 | 2.10 | 0.28 | 0.33 | 0.96 |
| **B8A5** | 0.52 | 0.61 | 1.07 | 0.28 | 0.36 | 0.59 |
| **B9A1** | 0.43 | 0.46 | 0.65 | 0.50 | 0.45 | 0.76 |
| **B9A2** | 0.38 | 0.68 | 1.04 | 0.51 | 0.43 | 0.33 |
| **B9A3** | 0.41 | 0.42 | 0.61 | - | - | - |
| **B9A4** | 0.25 | 0.25 | 0.64 | 0.55 | 0.55 | 0.48 |
| **B9A5** | 1.99 | 1.93 | 1.68 | 0.52 | 0.44 | 0.47 |
| **B10A1** | 0.55 | 0.72 | 0.64 | 0.57 | 0.51 | 0.49 |
| **B10A2** | 0.33 | 0.35 | 0.78 | 0.58 | 0.52 | 0.43 |
| **B10A3** | 0.42 | 0.43 | 0.71 | 0.53 | 0.52 | 0.53 |
| **B10A4** | 0.68 | 0.67 | 0.61 | 0.25 | 0.26 | 0.23 |
| **B10A5** | 1.14 | 1.14 | 1.19 | 0.26 | 0.31 | 0.23 |
| **Min** | 0.09 | 0.18 | 0.25 | 0.17 | 0.19 | 0.21 |
| **Max** | 1.99 | 1.93 | 2.10 | 0.78 | 1.43 | 2.19 |
| **Median** | 0.48 | 0.48 | 0.72 | 0.49 | 0.44 | 0.43 |

**ANNEX 6.** Indoor and outdoor $PM_1/PM_{10}$ and $PM_{2.5}/PM_{10}$ ratios

| | Indoor | | Outdoor | |
|---|---|---|---|---|
| | $PM_1/PM_{10}$ | $PM_{2.5}/PM_{10}$ | $PM_1/PM_{10}$ | $PM_{2.5}/PM_{10}$ |
| **B1A1** | 0.47 | 0.58 | 0.63 | 0.76 |
| **B1A2** | 0.42 | 0.52 | 0.61 | 0.75 |
| **B1A3** | 0.54 | 0.63 | 0.65 | 0.81 |
| **B1A4** | 0.47 | 0.57 | - | - |
| **B1A5** | 0.59 | 0.72 | 0.65 | 0.78 |
| **B2A1** | 0.56 | 0.70 | - | - |
| **B2A2** | 0.48 | 0.57 | 0.57 | 0.72 |
| **B2A3** | 0.22 | 0.33 | 0.58 | 0.73 |
| **B2A4** | 0.16 | 0.26 | 0.26 | 0.41 |
| **B2A5** | 0.26 | 0.40 | 0.24 | 0.40 |
| **B2A6** | 0.51 | 0.66 | 0.26 | 0.40 |
| **B3A1** | 0.55 | 0.68 | 0.66 | 0.84 |
| **B3A2** | 0.43 | 0.52 | 0.64 | 0.79 |
| **B3A3** | 0.65 | 0.76 | 0.75 | 0.91 |
| **B3A4** | 0.56 | 0.70 | 0.67 | 0.87 |
| **B4A1** | 0.78 | 0.91 | 0.53 | 0.73 |
| **B4A2** | 0.66 | 0.80 | 0.40 | 0.65 |
| **B4A3** | 0.60 | 0.79 | 0.40 | 0.64 |
| **B4A4** | 0.68 | 0.82 | 0.50 | 0.69 |
| **B4A5** | 0.51 | 0.65 | 0.53 | 0.73 |
| **B5A1** | 0.29 | 0.47 | 0.51 | 0.69 |
| **B5A2** | 0.48 | 0.62 | 0.55 | 0.75 |
| **B5A3** | 0.40 | 0.52 | - | - |
| **B5A4** | 0.30 | 0.41 | 0.66 | 0.78 |
| **B5A5** | 0.66 | 0.76 | - | - |
| **B6A1** | 0.31 | 0.44 | 0.52 | 0.68 |
| **B6A2** | 0.37 | 0.48 | 0.42 | 0.58 |
| **B6A3** | 0.38 | 0.52 | - | - |
| **B6A4** | 0.73 | 0.82 | 0.56 | 0.73 |
| **B6A5** | 0.77 | 0.88 | 0.40 | 0.58 |
| **B7A1** | 0.47 | 0.59 | 0.44 | 0.55 |
| **B7A2** | 0.39 | 0.47 | 0.32 | 0.42 |
| **B7A3** | 0.40 | 0.55 | 0.34 | 0.45 |
| **B7A4** | 0.74 | 0.81 | 0.42 | 0.53 |

**Annex 6.** Continued

|  | Indoor | | Outdoor | |
|---|---|---|---|---|
|  | $PM_1/PM_{10}$ | $PM_{2.5}/PM_{10}$ | $PM_1/PM_{10}$ | $PM_{2.5}/PM_{10}$ |
| **B7A5** | 0.53 | 0.64 | 0.43 | 0.53 |
| **B8A1** | 0.35 | 0.45 | 0.59 | 0.70 |
| **B8A2** | 0.29 | 0.45 | 0.57 | 0.72 |
| **B8A3** | 0.38 | 0.55 | 0.57 | 0.70 |
| **B8A4** | 0.24 | 0.38 | 0.28 | 0.41 |
| **B8A5** | 0.34 | 0.50 | 0.47 | 0.61 |
| **B9A1** | 0.10 | 0.19 | 0.30 | 0.39 |
| **B9A2** | 0.28 | 0.43 | 0.38 | 0.51 |
| **B9A3** | 0.18 | 0.27 | - | - |
| **B9A4** | 0.29 | 0.38 | 0.26 | 0.40 |
| **B9A5** | 0.30 | 0.42 | 0.22 | 0.34 |
| **B10A1** | 0.34 | 0.51 | 0.24 | 0.34 |
| **B10A2** | 0.31 | 0.41 | 0.28 | 0.39 |
| **B10A3** | 0.21 | 0.32 | 0.22 | 0.35 |
| **B10A4** | 0.58 | 0.83 | 0.52 | 0.64 |
| **B10A5** | 0.63 | 0.73 | 0.48 | 0.65 |
| **Min** | 0.10 | 0.19 | 0.22 | 0.34 |
| **Max** | 0.78 | 0.91 | 0.75 | 0.91 |
| **Median** | 0.42 | 0.55 | 0.49 | 0.65 |

**ANNEX 7.** I/O ratios of $PM_1$ (A) and $PM_{10}$ (B)
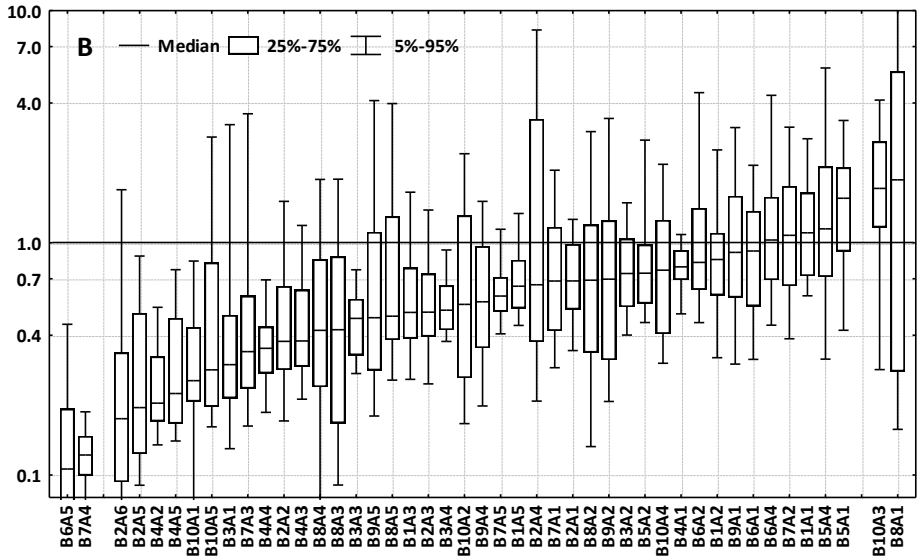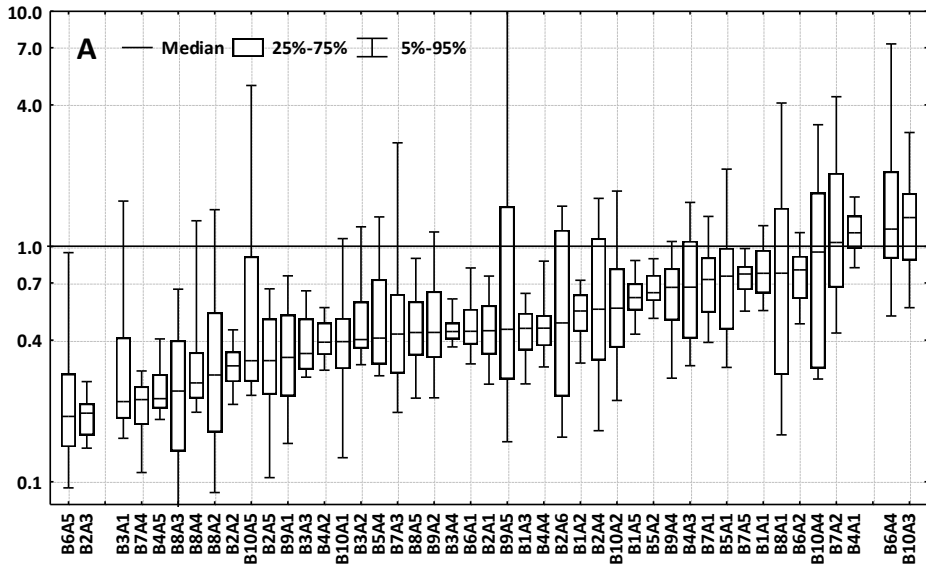
**ANNEX 8.** Nighttime (background) PM$_{10}$ levels
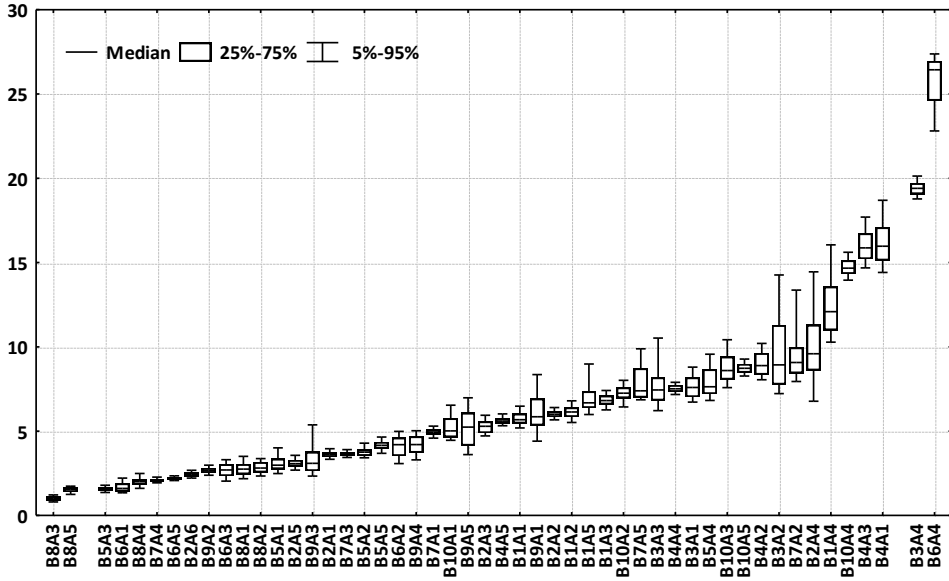
**ANNEX 9.** Outdoor temperature and relative humidity during the PM sampling period (Dec 2011 - Dec 2012). Average and standard deviation values are shown by month

| Year/Month | Temperature, °C | Relative humidity, % |
|:---:|:---:|:---:|
| **2011/12** | 1.80 (2.80) | 88.32 (5.17) |
| **2012/01** | -2.98 (5.65) | 83.66 (9.46) |
| **2012/02** | -9.04 (8.91) | 79.63 (9.41) |
| **2012/03** | 1.93 (3.97) | 76.35 (15.44) |
| **2012/04** | 7.74 (6.65) | 66.42 (21.24) |
| **2012/10** | 7.36 (4.53) | 86.34 (9.88) |
| **2012/11** | 4.82 (2.09) | 89.12 (7.43) |
| **2012/12** | -4.28 (4.72) | 81.92 (7.99) |

**ANNEX 10.** Living environment and building information from the housing questionnaires and diaries (prevalence values + 95 % confidence intervals)

| Living environment and building information | Value, % |
|---|---|
| **Facilities** | |
| Central heating | 87.5 (78.8-96.2) |
| Mechanical exhaust ventilation (restroom fan) | 35.7 (23.2-48.3) |
| Mechanical support ventilation | 14.3 (5.1-23.5) |
| Fresh air vents in bedrooms | 30.4 (18.3-42.4) |
| Air humidifier | 3.6 (-1.3-8.4) |
| Air purifier | 5.4 (-0.5-11.3) |
| Gas stove | 83.9 (74.3-93.5) |
| Fireplace/wood burning oven | 3.6 (-1.3-8.4) |
| Kitchen vent hood | 75.0 (63.7-86.3) |
| **Windows** | |
| Single pane | 1.9 (-1.8-5.7) |
| Double pane | 78.8 (67.7-89.9) |
| Triple pane | 13.5 (4.2-22.7) |
| **Window open in winter daily/almost daily** | |
| Kitchen | 68.5 (56.1-80.9) |
| Bedroom(s) | 72.5 (60.3-84.8) |
| Living room | 54.9 (41.2-68.6) |
| Bathroom | 54.5 (25.1-84.0) |
| Other area | 33.3 (-20.0-86.7) |
| **Temperature conditions in winter** | |
| Suitably warm | 33.9 (21.5-46.3) |
| Too cold | 37.5 (24.8-50.2) |
| Too warm | 7.1 (0.4-13.9) |
| Draughty | 3.6 (-1.3-8.4) |
| Cold floor surfaces, etc. | 32.1 (19.9-44.4) |
| **Moisture condensation on windows in winter** | |
| Daily/almost daily | 37.0 (24.2-49.9) |
| Weekly | 13.0 (4.0-21.9) |
| Less frequently | 35.2 (22.4-47.9) |
| Never | 14.8 (5.3-24.3) |