# ELDERLY EXPOSURE TO AIR POLLUTANTS

Measuring, assessing and modelling

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Measuring, assessing and modelling

#### **Proefschrift**

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### The ageing process is of course a biological reality which has its own dynamic, largely beyond human control.

(Gorman M. Development and the rights of older people. In: Randel J, et al., Eds. The ageing and development report: poverty, independence and the world's older people. London, Earthscan Publications Ltd.,1999: 3-21)

Nevertheless, it is our responsibility to provide the suitable tools to keep this process less painful as possible.

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#### CHAPTER I. INTRODUCTION

#### 1.1 Elderly population – an emerging concern

The concerns regarding air pollution effects in elderly population started to appear in 1998, after A. Davies and G. Viegi had been invited by M. Decramer and R. Loddenkemper (on behalf of European Respiratory Society) to submit a scientific project regarding the topic. Due to the scarcity of knowledge about air pollution effects in elderly population, this susceptible group was selected to be the central key of the project AFORDEE (Anticipation of Focus On Respiratory Disease in the European Elderly) As deliverable of AFORDEE, a workshop named "Air Pollution Effects in the Elderly" was held in Pisa in 2001, resulting 16 articles published in European Respiratory Journal (2003, Suppl. 40).

Before this, APHEA Project (Air Pollution and Health: A European Approach), that had started in 1993, had investigated the short-term effects of air pollution on health. Beyond this project, many other studies have dedicated their efforts on relation with air pollutants and morbidity/mortality (Pope et al., 2002; Simoni et al., 2003; Simkhovich et al., 2008; Tena & Clarà, 2012; Almeida et al., 2014a, Cruz et al., 2015). Even though many of these works have been conducted in the general population, any focus their attention on elderly specifically. GERIE was the first study that used different sources of information in relation to adverse health reactions of environmental hazards in elderly, taking into account indoor environments (Bentayeb et al., 2014). This pioneer project had demonstrated independent effects of several indoor air pollutants and comfort parameters on respiratory morbidity, being greater in the case of poor ventilation and in those aged > 80 years. Old people spend the majority of their life in indoor environments, especially those who are institutionalized in Elderly Care Centers. These places starting to have an important role in developed societies since life expectancy had increased more than 5 years in last 30 years. In Portugal, the number of Elderly Care Centers increased 49% between 1998 and 2010 (GEP/MSSS, 2010). These places may present high concentrations of several air pollutants and it is known that the more intense and longer is the exposure, the greater may be the risk.

The older population is growing faster than the total population in practically all regions of the world – and the difference in growth rates is increasing (United Nations, 2012). Since 1996 until 2008 the number of adults aged > 65 years increased 31% (from 380 million to 500 million). According to the United Nations (2013) the percentage of total population aged 60 years or over in the world was 11% for the year 2010 and is estimated to be 18% for 2050. Since 1950, the world is assisting an inversion of age pyramid, with a constant increase on number of elders worldwide (Figure 1.1). In 2050 the proportion of older persons will be double than children and this growth tends to be more significant in developed countries. The ageing index in Europe will be 263 per hundred children under 15 years old in 2050, remaining the highest index throughout the other continents. Europe presents the highest percentages of old people worldwide, being Africa the continent with the lowest percentage of population with more than 60 years. Since 2009, Portugal is the 4<sup>th</sup>

oldest country in Europe (EU 28), with almost 20% of population with more than 60 years old (Eurostat, 2014). The increase on life expectancy is directly correlated with chronic diseases that affect elderly individuals, such as cardiopulmonary diseases, cancers, diabetes and kidney failure (Bentayeb et al., 2013).

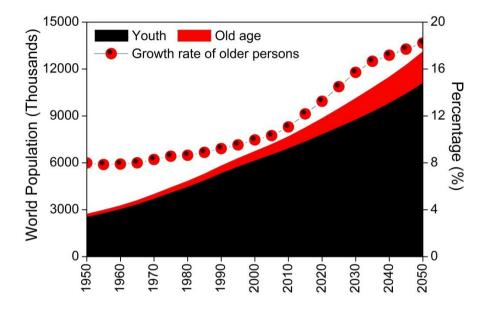


Figure 1.1 – Distribution of population by youth and older persons and growth rate of old people (values obtained in United Nations reports [2011 and 2013]).

Air pollution exposure has been shown to be linked to respiratory and cardiovascular diseases (WHO, 2013). It is estimated that approximately 7 million premature deaths are linked to air pollution world-wile (WHO, 2014). The vast majority of air pollution deaths are due to cardiovascular diseases: 40% related to ischaemic heart disease; 40% stroke; 11% chronic obstructive pulmonary disease (COPD) (WHO, 2014). This kind of human effects may be significantly worse when susceptible population are considered, such as elders.

#### 1.2 Indoor air quality

The concerning about exposure to air pollution and their possible human health effects is not a current problem. Actually, in 1875 arose the first legal document containing a section called "nuisances" where it was required the decrease of smoke pollution in urban areas (Public Health Act, 1875). Meanwhile, several epidemiological studies have established

associations between exposure to air pollution and adverse human health effects (Almeida et al, 2014a; Pope et al., 2011, 2002). However, epidemiological associations between air pollutants and health outcomes are based predominantly on ambient air measurements where it is assumed that all people in a given region have the same exposure level, which is often obtained from a few air quality monitors and reflects the entire community. Nevertheless, poor correlations have been found between ambient air pollutants' concentrations and personal exposure to these air pollutants (Meng et al., 2005) because actual exposure is strongly related to the individual time activity patterns, followed by its distance from each pollutant source.

Although indoor concentration and number of carcinogenic air pollutants has been decreased since the 1950s (Weschler, 2009), all of the previously evidences, plus the changes on life-style and the fact that people spend a large part of their life inside the indoor environments (from 80 to 90%), have promoted an increase on exposure to indoor air pollutants (Byčenkienė et al., 2009; Zhao, et al., 2009; Dales et al., 2008; Leech et al., 2002; Klepeis, et al., 2001). Consequently, we are witnessing an intensification of studies developed by the scientific community concerning Indoor Air Quality (IAQ) and its effects upon health (Canha et al., 2012a; Franck et al., 2011; Almeida et al., 2011; WHO, 2010; Saliba et al., 2009; Fraga et al., 2008; Fromme et al., 2007; Kosonen, 2004; Lee et al., 2002; Wilson, 1996; Allen & Miguel, 1995).

Indoor air pollution is caused by a combination of several factors: hazardous substances that are emitted from the outdoors, buildings, construction materials, furnishings, equipment, inadequate ventilation, indoors human activities, etc. (Canha et al., 2013; Pegas et al., 2011a,b; Canha et al., 2010; Viegas et al., 2010; Weschler, 2009). Physical factors such as air temperature, air velocity and relative humidity are usually used as indicators of thermal comfort, in IAQ studies. The main chemical parameters used to characterize the IAQ are carbon monoxide (CO) and dioxide (CO<sub>2</sub>), the volatile organic compounds (VOC), the formaldehyde ( $H_2$ CO), the ozone ( $O_3$ ) and, the particulate matter (PM).

Carbon dioxide is a pollutant emitted by the human metabolism and is commonly used as an indicator of occupancy and poor ventilation (Hänninen, 2013). Since the  $CO_2$  is associated with the human occupancy,  $CO_2$  can be measured in indoor environments as an indicator of air quality (Lee et. al., 2002).

The VOCs and the formaldehyde are also associated with indoor sources. According to Jantunen (2007) high indoor concentrations of those pollutants rarely are originated from outdoor air. Usually, VOCs and the formaldehyde are released from indoor materials, such as 1) office furniture; 2) cabinetry; 3) carpet title; 4) vinyl wall coverings; 5) paints; 6) adhesives; 7) glue; 8) varnish; and 9) cleaning products (Weschler, 2009; Destaillats et. al., 2008; Valuntaitè & Girgždienė, 2008; Bernstein et. al., 2008).

Another important source of indoor pollutants is the electronic equipment. Computers, copiers and printers are characterized as indoor sources of VOCs, O<sub>3</sub> and particles (Weschler, 2009; Destaillats et. al., 2008; Valuntaitė & Girgždienė, 2008). Ozone can also be provided from outdoor sources and air purifiers (Bernstein et. al., 2008).

Indoor particle concentration depends a lot on the penetration of outdoor particles into the indoor environment and on the intensity of indoor aerosol sources (Estoková et. al., 2010). Inhaled particles penetrate into the respiratory tract where they target different anatomical sites, depending among other properties on the aerodynamic size (Figure 1.2). PM10 is the particulate matter with an aerodynamic diameter (AD) lower than 10  $\mu$ m and it is also called as thoracic particles. On the other hand, PM2.5 (AD,  $\leq$ 2.5  $\mu$ m) is named as fine fraction. While the particles with an AD higher than 10  $\mu$ m tends to be hold in the first barriers (upper airways), the PM2.5 can achieve the lower airways. As lower is the particle size, deeper it reach into respiratory system. Ultra-fine particles and nanoparticles can go deep on respiratory system and, consequently, its components can be absorbed by the alveoli and be translocated to other organs.

Despite the fact that PM is one of the chemical parameters used to characterize the indoor air quality, since the majority of national and international guidelines consider PM10 as a monitored indoor air pollutant, among other chemical and physical pollutants (Portaria 353-A/2013; HKEPD, 2003; HN 35:2002; EPA, 2000), there is a special challenge concerning on interpretation of its results. Due to this issue, some researchers have investigated which properties of ambient aerosol are responsible for health effects; whether certain particulate chemical components are more harmful than others (Suh et al., 2011; Zanobeti et al., 2009); and the particle size as an important determinant of the site and efficiency of pulmonary deposition (Anderson et al., 2008).

According to Morawska (2013), up to 30% of the burden of disease from PM exposure can be attributed to indoor-generated particles, signifying that indoor environments are likely to be a dominant factor affecting human health. This initiated a debate as to whether ambient PM is a good surrogate for exposure to PM once the composition and toxicity of indoor PM is very complex, with similarities but also differences to outdoor aerosols. Therefore, personal integrated exposure to PM components is of considerable importance as it is the key determinant of the PM dose received by an individual and thus directly influences the health impacts.

More recently, researchers started to be concerned with the exposure to nanoparticles that takes place essentially indoors, at home, in school, or at the workplace and depending on the amount of time that an individual spends in areas with high or low concentrations (Coelho et al., 2005). Therefore, it is expected an increase of exposure to nanoparticles as a result of an increase in production and use of engineering nano-materials (Asbach et al., 2009). Numerous studies have already showed that airborne nanoparticles have a potential to evoke serious adverse human health effects when deposited in the respiratory tract (Oberdörster et al. 2005). The most important part of the lung is the alveolar region, with their enormous surface areas, that increase the possibility to transfer nanoparticles into the blood stream and from thereon into all end organs of the body. Other potential consequence is the oxidative stress in the body which can occur due to a typical indoor exposure to nanoparticles (Weichenthal et al., 2007; Vinzents et al., 2005). Among indoor physical and chemical pollutants, it is necessary to consider bioaerosols that consist of airborne particles

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that either contain living organisms such as bacteria, viruses and fungi or originate from living organisms. Bioaerosols are ubiquitous, highly variable, complex, natural or synthetic in origin and contribute to approximately 5-34% of indoor air pollution (Srikanth et al., 2008; Bio-aerosols, 2007). Several studies positively correlated indoor exposure to microorganisms and microbial components with adverse health effects including headache and respiratory symptoms (Douwes et al., 2003). Considering specifically fungi, their spores are complex agents that may contain multiple hazardous components. Health hazards may differ across species because fungi may produce different allergens and mycotoxins. Moreover, some species also infect humans (Eduard & Halstensen, 2009). Most infections occur in immunocompromised hosts or as a secondary infection, following inhalation of fungal spores or the toxins produced by them (Srikanth et al., 2008).

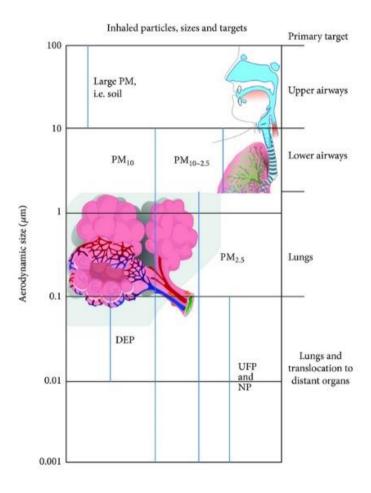


Figure 1.2 – Schematization of the size and main target for particles (From Nemmar et al., 2013).

#### 1.3 Exposure and dose to air pollutants

Exposure is defined as "an event that occurs when there is a contact at a boundary between a human and the environment with a contaminant of a specific concentration for an interval of time" (NAS, 1991).

According to several authors there are three methodologies that can be used to measure or estimate the exposure to airborne pollutants: 1) the direct method; 2) the indirect method; and 3) the biological marker method (Trasande & Landrigan, 2004; Moschandreas & Saksena, 2002; National Academy of Sciences (NAS), 1991). In the direct method, human exposure is measured in real-time by portable instruments, used by volunteers, which record the air pollutants concentrations near the breathing zone. This approach is very relevant since researchers showed the existence of a phenomenon named Personal Cloud due to the fact that pollutants concentration measured using personal monitors were consistently higher than those measured by a stationary monitor. A personal cloud occurs, at least in part, because indoor PM sources are usually associated with personal activities, which results in elevated concentrations near people. In fact, one of the scientific explanations of this discrepancy is a source proximity effect, in which pollutant sources close to the person cause elevated and highly variable exposures (McBride et al., 1999). Other hypotheses include the possible role of electrostatic charges (Schneider et al., 1993). One of the biggest advantages of using personal monitors is the fact that this accurately reflects the personal exposure to key air pollutants. This is because the estimated personal exposure by using personal monitors is more relevant that the data collected from stationary ambient monitoring equipment, being the latest poorly correlated with total personal exposure. Personal exposure can be affected by indoor PM concentration (which is influenced by different kind of sources, air exchange rates, etc.), commuting and secondhand smoke (Rojas-Bracho et al., 2004). All of these issues plus the fact that people spend the majority of their time indoors, visiting a variety of micro-environments with many different PM sources, may justify the poor correlation of PM ambient concentrations measured by fixed stationary monitors and total personal exposure (Steinle et al., 2013). Nevertheless, to keep and carry instruments while moving around the volunteers become to be annoyed, disturbed and stressed. Plus, their prohibitive cost in large study populations unfeasible any work.

So, an indirect approach can be used as an alternative method that estimates the exposure by integrating the time that people spend in each micro-environment and the concentration of the pollutants for the period of interest, (ILO, UNep & WHO 2000; Sexton, Callahan & Bryan, 1995), as is explained by Equation 1.1:

$$E_{i} = \frac{\sum_{j=1}^{m} c_{ij} \cdot t_{ij}}{\sum_{j=1}^{m} t_{ij}}$$
(1.1)

Where  $C_{ij}$  is the concentration of the pollutant measured in the  $j^{th}$  micro-environment of the  $i^{th}$  individual,  $t_{ij}$  is the time spent by the  $i^{th}$  individual in the  $j^{th}$  micro-environment. The total number of micro-environments is m such that:

$$\sum_{i=1}^{m} t_{ij} = 24 h$$

To know the amount of time that people spend in different micro-environments it is necessary to use information obtained by questionnaires (Freeman & Tejada, 2002). The three main outputs of the application of questionnaires are: 1) average time of each type of activity during the day for all people; 2) percentage of people who participated in a given activity on the selected day; 3) average of time spent on the activity by those who actually participated in it on the given day (Andorka, 1987).

There are two different theories to assess the dose, since in one case it is possible to calculate the inhaled dose and in the other the deposited dose. According to the first theory it is crucial to integrate the time spent in each micro-environment (t), the concentration of the pollutants for the period of interest (C), the inhalation rate (IR) and the body weight (BW) according to the Equation 1.2:

$$E_{di} = \frac{\sum_{j=1}^{m} (c_{ij} \cdot t_{ij} \cdot IR_{ij})}{\sum_{j=1}^{m} t_{ij}}$$
(1.2)

The IR's used for the three different micro-environments – bedroom, living-room and outdoor – were recommended by USEPA (2011) for people with more than 61 years old in three distinct activities – sleep, sedentary and light intensity, respectively. These values were selected to be used as the recommended inhalation rates since they were based on three studies: USEPA (2009), Stifelman (2007) and Brochu et al. (2006). The body weight used was 80 kg, also based on USEPA (2011). This methodology was performed to

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calculate the inhaled dose of the PM components, such as carbonaceous fraction and trace elements (Almeida-Silva et al., 2015). In the second theory, the deposited dose can be calculated according to a numerical model, which gives the dose in each part of the human respiratory tract; the extrathoracic and each generation of the lungs. The model uses the particles concentration at the breathing zone as input and takes into account the time spent by people in each micro-environment, the physicochemical characteristics of the PM and the physiological parameters of the people exposed in order to calculate the dose. This model was already descripted and validated in several publications elsewhere (Mitsakou et al., 2005; Mitsakou et al., 2007a,b). The greatest difference between the two methodologies is that the first is an empirical approach, thus limited in the parameters range of its development, while the second numerical approach is mechanistic, i.e. based on laws of physics, thus its applicability is much wider. Moreover, the model gives a detailed description of the dose of particles in HRT, since the model estimates the PM deposition in all different regions of the respiratory tract. In order to summarise part of the methodology implemented in this thesis the Figure 1.3 was created. From exposure to deposited dose several steps were done in order to fulfil one of the goals of this thesis.

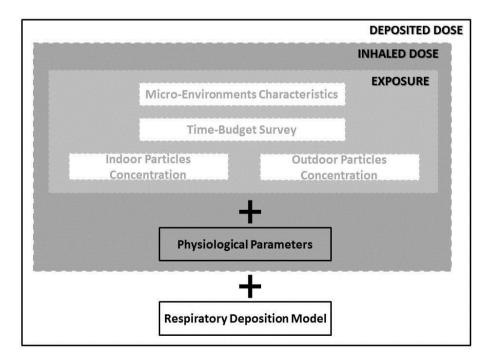


Figure 1.3. Schematic diagram illustrating the parameters for calculating the exposure, inhaled and deposited dose in respiratory tract.

#### 1.4 Risk assessment paradigm

One of the first designations of risk assessment was developed by National Research Council of National Academy of Sciences in 1983, and was defined by "the characterization of the potential adverse health effects of human exposures to environmental hazards" (NAS, 1983). This can also be defined as a formalized basis for the objective evaluation of risk in a manner in which assumptions and uncertainties are clearly considered and presented.

Historically, the risk assessment process was carried out in a six-step sequence: 1) emission sources; 2) transport; 3) hazard identification and quantification; 4) dose; 5) dose response; and 6) risk. Though, this scheme was constrained by two limitations. Firstly, individuals and the population exposed were essentially ignored. Moreover, risk assessment requires knowledge of the number of individuals exposed to the estimated level and this evidence was not taken into account by the previous risk assessment process. Secondly, the six-step sequence ignored indoor sources, indoor environments and their pollutant emissions where in some cases is likely to be a substantive portion of exposure to pollutants and, for certain pollutants, the largest portion.

Consequently, risk assessment paradigm flows in a logical, in order to introduce characteristics of a population and estimated of pollutants levels at the population site(s). Therefore, a scientific paradigm shifts to a stepwise fashion that includes the following steps: 1) the evaluation of emission sources; 2) the identification and quantification of hazards; 3) the exposure assessment; 4) the quantification of the dose; and 5) the study of effects on human health (Figure 1.4). The last step can also be defined as risk characterization, being the culmination of the other steps. Hazard, exposure and dose assessment are considered in juxtaposition to determine risk or to determine what additional data are needed to calculate risk or to refine risk estimates.

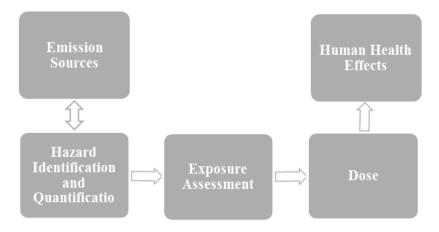


Figure 1.4 – Risk Assessment Paradigm applied in this work.

The ultimate purpose of a risk assessment paradigm is to help the organization/decision maker/researcher make good decisions to achieve their goals. This is always a cyclical process: the last activity is to return to the first step (Kallman and Maric, 2004). This means that this is an interactive and dynamic process, with constants reviews, updates and achievements.

#### 1.5 Objectives and Thesis outline

The present study aims to estimate the human exposure to air pollutants, given special attention to one of the most susceptible population – elders. To fulfil the goal, the work was conducted follow the risk assessment paradigm, described previously, and, consequently, divided into 5 tasks:

- Characterization of the IAQ in ECCs, evaluating an orchestra of indoor physical, chemical and biological pollutants;
- Assessment of daily integrated exposure and inhaled dose to air pollutants;
- Determination of elderly personal daily exposure;
- Estimation of PM deposited dose in elderly respiratory tract by modelling;
- Source apportionment of indoor PM in an Elderly Care Center.

All of this work involved the collaboration with several institutions and organisms. Major of Loures city (Câmara Municipal de Loures, Portugal), by the Department of Social Cohesion and Habitation, gave the assistance to directly contact the Elderly Care Centers; Lisbon School of Health Technology (ESTeSL, Portugal), by the Department of Environmental Health, provided a set of equipments and helped with the identification and quantification of fungi contamination; Aveiro University (UA, Portugal), by Centre for Environmental and Marine Studies (CESAM), made available a set of equipments and analysed the PM carbonaceous components; and, finally, National Centre for Scientific Research "DEMOKRITOS" gave the assistance to estimate the PM deposited dose in elderly lungs by modelling and also analysed the PM ions and carbonaceous components in order to perform a source apportionment.

Chapter 2 describes the characteristics of the selected Elderly Care Centers and its institutionalized elders. In this chapter an orchestra of indoor physical, chemical and biological pollutants was assessed, as well as, the time-budget survey of the studied population, in order to calculate the daily average exposure and the daily average inhaled dose. This work showed that besides living in the same area, the exposure and the inhaled dose of the studied elders differed significantly. Moreover, the work demonstrated that an accurate measurement of integrated exposure is essential to provide an adequate evaluation of the particles dose-response relation.

In Chapter 3 inhaled dose of particles was calculated for autonomous and institutionalized elders. Autonomous elders carried out a real-time PM monitor during four 24 h measuring campaigns and fulfilled an activity diary, simultaneously. Subsequently, the PM deposited

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dose on elderly respiratory tract was estimated by modelling. Moreover, the same mathematical model was applied to calculate the PM deposited dose in respiratory tract of the elders studied in Chapter 2.

Chapter 4 presents the results of the indoor PM source apportionment made in an Elderly Care Center. For those two 2-weeks sampling campaigns were conducted to collect PM10. The elemental composition, the organic and elemental carbon and the ions were determined in order to identify emission sources.

### CHAPTER II. ELDERLY EXPOSURE TO INDOOR AIR AEROSOLS

This chapter is a version based on the following publications: 1) Almeida-Silva M, Wolterbeek HT and Almeida SM, 2014. Elderly exposure to indoor air pollutants. *Atmos Environ*, 84: 54-63; 2) Viegas C, Almeida-Silva M, Gomes AQ, Wolterbeek HT and Almeida SM, 2014. Fungal contamination assessment in Portuguese elderly care centres. *J Toxicol Env Heal A*, 77 (1-3): 14-23; 3) Almeida-Silva M, Almeida SM, Gomes JF, Albuquerque PC and Wolterbeek HT, 2014. Determination of airborne nanoparticles in elderly care centers. *J Toxicol Env Heal A*, 77 (14-16): 867-878; 4) Almeida-Silva M, Almeida SM, Pegas PN, Nunes, T, Alves, CA and Wolterbeek HT, 2015. Exposure and dose assessment to particle components among an elderly population. *Atmos Environ*, 102: 156-166.

#### 2.1 Abstract

The aim of this work was to fully characterize the indoor air quality in Elderly Care Centers (ECCs) in order to assess the elders' daily exposure and the inhaled dose to air pollutants. Ten ECCs hosting 384 elders were selected in Lisbon and Loures, Portugal. Firstly, a timebudget survey was created based on questionnaires applied in the studied sites. Secondly, a set of physical, chemical and biological parameters were measured during the occupancy period in the two micro-environments where elders spend most of their time: bedroom and living-room. Finally, daily exposure was calculated by integrating the time spend in each micro-environment and the concentration of the pollutants for the period of interest. This parameter, together with the inhalation rate and the body weight, were used to calculate the daily inhaled dose. Results showed that elders spend 95% of their time indoors, splitted between bedrooms and living-rooms. In general, living-rooms presented highest pollutants concentrations, with exception for CO<sub>2</sub> and CO. Results showed that the PM10 indoor concentrations did not exceed the national and the international limit values and that PM10 concentration in living-rooms were significantly higher than in bedrooms. Zn and Cr presented higher concentrations in the indoor environments indicating the existence of indoor sources for these elements. The most enriched elements in relation to a reference soil were Sb, Zn, As and Cr indicating their association with anthropogenic sources. Results also showed that besides living in the same area, the exposure and the inhaled dose of the studied elders differed significantly. Penicillium sp. was the most frequent isolated (38.1%), followed by Aspergillus sp. (16.3%) and Chrysonilia sp. (4.2%). The living-room was the indoor micro-environment with lowest fungal concentration and the storage area was highest. PM10 daily exposure and daily inhaled dose ranged between 11 – 16 µg.m<sup>-3</sup> and  $20x10^{-3} - 28x10^{-3}$  µg.kg<sup>-1</sup>, respectively. Deposited surface area of nanoparticles were also assessed and ranged between 10 µm<sup>2</sup>.cm<sup>-3</sup> and 46 µm<sup>2</sup>.cm<sup>-3</sup>. Plus, bedrooms were the micro-environment that most contributed to the PM10 exposure and inhaled dose. This approach allows the identification of the micro-environments with highest impacts on elderly exposure and proved to be an essential tool to identify health risks, set and review air quality standards and evaluate effective policy interventions.

#### 2.2 Introduction

Several epidemiological studies have established associations between exposure to Particulate Matter (PM) and adverse human health effects (Almeida et al, 2014a; Pope et al., 2011, 2002). More recently, some researchers have investigated which properties of ambient aerosol are responsible for health effects; whether certain particulate chemical components are more harmful than others (Suh et al., 2011; Zanobetti et al., 2009); and the particle size as an important determinant of the site and efficiency of pulmonary deposition (Anderson et al., 2008). Nevertheless, poor correlations have been found between ambient PM concentrations and personal exposure to PM (Meng et al., 2005) because actual exposure is strongly related to the individual time activity patterns, followed by its distance from each particle source.

We are observing an increase of studies developed by the scientific community concerning Indoor Air Quality (IAQ) and its effects upon health, (Canha et al., 2012a; Franck et al., 2011; Almeida et al., 2011; WHO, 2010; Canha et al., 2010; Saliba et al., 2009; Fraga et al., 2008; Fromme et al., 2007; Kosonen, 2004; Lee et al., 2002; Spengler, 1996; Allen & Miguel, 1995), since people spend a large part of their life inside the indoor environments (more than 80-90%) which have promoted an increase on exposure to indoor air pollutants (Byčenkienė et al., 2009; Zhao, et al., 2009; Dales et al., 2008; Leech et al., 2002; Klepeis, et al., 2001).

The effective integrated exposure assessment should be estimated by the time spent by people in different environments and the concentration of the pollutants for the period of interest (ILO, UNep & WHO, 2000; Sexton, Callahan & Bryan 1995). To assess the daily inhaled dose is crucial to integrate the time spend in each micro-environment, the concentration of the pollutants for the period of interest, the inhalation rate and the body weight. The use of this both approaches is useful not only to provide an adequate evaluation of the pollutants dose-response relation, but also to identify health risks, set and review air quality standards and assess effective policy interventions.

These facts are particularly relevant when we are talking about institutionalized elderly people not only because they are considered a susceptible group but also because they spend the majority of their time indoors (Almeida-Silva et al., 2014a; Prasad et al., 2003). Europe presents the highest percentages of old people worldwide, being Africa the continent with the lowest percentage of population with more than 60 years (United Nations, 2012). Portugal is the 4<sup>th</sup> oldest country in Europe (EU 28), with almost 20% of population with more than 60 years old (Eurostat, 2014). In Portugal, the number of Elderly Care Centers (ECCs) increased 49% between 1998 and 2010 (GEP/MSSS, 2010). Despite

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the importance of healthy air in ECCs, IAQ studies have been focused principally on schools (e.g. Canha et al., 2014a; Canha et al., 2013; Canha et al., 2012a; Pegas et al., 2011a,b; Canha et al., 2011; Pegas et al., 2010; Canha et al., 2010); homes (e.g. Osman et al., 2007) and offices (e.g. Bluyssen et al., 1996). Furthermore, as far as we know the daily exposure to carbon dioxide, carbon monoxide, PM in different sizes fractions, total volatile organic compounds and PM components (trace elements and carbonaceous components) and the estimation of the daily dose to PM components (trace elements and carbonaceous components) has never been done before, even for the children which are the most studied population. GERIE was the first research work that studied the air quality in ECCs, explaining health and environmental disparities in elderly in the European Union. According to the GERIE results more and detailed studies are needed to better characterize this population and its exposure to air pollutants and to seek to identify best practices.

The objective of this work was to follow the risk assessment paradigm in order to 1) characterize the IAQ in ECCs, evaluating an orchestra of indoor physical, chemical and biological pollutants; 2) assess the daily integrated exposure to different air pollutants; and 3) calculate the inhaled dose of institutionalized elders.

#### 2.3 Material and methods

The current study was carried out in 10 ECCs, located in Lisbon and Loures, District of Lisbon (Figure 2.1). This region is located in the west of Portugal, on the Atlantic Ocean coast, being the westernmost capital in Europe. The metropolitan area of Lisbon has an area of 2870 km<sup>2</sup> and has almost 3 million inhabitants. Loures is one of the 18 regions that belongs to the metropolitan area of Lisbon, having around 205 thousands inhabitants in 160 km<sup>2</sup> of area (INE, 2012).

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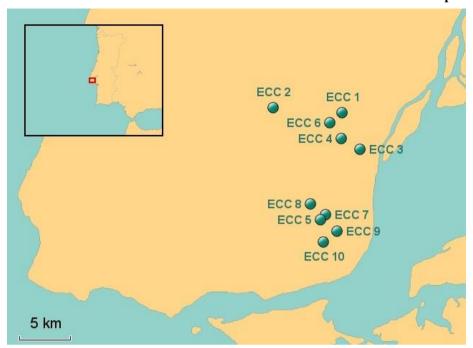


Figure 2.1 – Geographical distribution of the 10 ECCs selected.

#### 2.3.1 Characterization of population

The present work was developed in collaboration with 384 old people living in ECCs which had a range of 7-95 occupants per institution. Table 2.1 shows the characterization of the studied population. Women not only were presented in higher number, but also were older than men and were the ones who were bedridden in greater number (6% of the studied population were bedridden women).

Table 2.1 – Characterization of the studied population. The results are presented in absolute values (N/A: Not Applicable).

	Wome	n	Men	
	N	Age (min-max)	N	Age (min-max)
ECC 1	26	84 (68-99)	11	81 (67-91)
ECC 2	40	87 (74-99)	11	88 (76-96)
ECC 3	39	88 (77-100)	0	N/A
ECC 4	42	84 (70-99)	24	82 (70-90)
ECC 5	9	85 (72-98)	0	N/A
ECC 6	55	81 (65-96)	40	78 (65-95)
ECC 7	5	87 (82-95)	4	82 (80-82)
ECC 8	51	80 (65-96)	0	N/A
ECC 9	6	86 (67-101)	1	86
ECC 10	20	91 (69-104)	0	N/A
Total	293 (2	2 bedridden)	91 (6 t	pedridden)

#### 2.3.2 Characterization of 10 ECCs

Considering the particular characteristics of the surrounding environment, ECCs were classified as urban or sub-urban. Table 2.2 shows that four ECCs were located in a sub-urban area while six ECCs were placed in an urban area. A technical questionnaire was applied in order to characterize the buildings. This questionnaire included information about: 1) ventilation systems; 2) types of indoor materials; 3) ventilation and cleaning practices; 4) type of building construction; 5) thermal isolation of the building and 6) characterization of the building envelope. A resume of the information obtained from the application of this questionnaire is presented in Table 2.2.

Table 2.2 – Characterization of Elderly Care Centers.

ECC	Season of sampling	Zone	Type of building	N.º of Beds	HVAC	Pavement		Pavement Windows		Cleaning frequency		
	Samping		building	Deus		BR	LR	BR	LR	BR	LR	
ECC 1	Autumn	Sub- urban	Villa (with 3 floors)	37	Yes	Vinyl	Ероху		ole glass minum	1xday		
ECC 2	Autumn	Sub- urban	Villa (with 3 floors)	51	No	Wo	Wood		Double glass aluminum		1xday	
ECC 3	Autumn	Sub- urban	Villa (with 2 floors)	40	No	Wo	Wood		Double glass aluminum		1xday	
ECC 4	Autumn	Urban	Villa (with 3 floors)	69	No	Vir	Vinyl		Vinyl Tile Double glass aluminum		1xc	day
ECC 5	Autumn	Urban	Villa (with 2 floors)	10	No	Wo	Wood		mple glass wood	1xc	day	
ECC 6	Autumn	Sub- urban	Building (with 6 floors)	131	No	Linol	Linoleum		mple glass lluminum	1xday	2xday	
ECC 7	Autumn	Urban	Villa (with 4 floors)	15	No	Wo	Wood		ole glass minum	1xc	day	
ECC 8	Winter	Urban	Villa (with 2 floors)	53	No	Vinyl			ole glass minum	1xweek	1xday	
ECC 9	Winter	Urban	Apartment on the 1 <sup>st</sup> floor of a building with 5 floors	15	No	Floating Wood			ole glass minum	1xday	2xday	
ECC 10	Winter	Urban	Apartment on the 3 <sup>rd</sup> floor of a building with 4 floors	23	No	Wood			ole glass minum	1xc	day	

#### 2.3.3 Time budget-survey

Time-budget surveys (TBS) are useful tools to estimate the people exposure to air pollutants, due to the fact that they give us the time spent by people on different locations.

A time-budget survey was built for 384 elders. For this a close-ended questionnaire was designed, which included information about different activities developed during the day, mealtimes, sleep times, micro-environments where they spent their time, etc. The questionnaire differentiated between time allocation on weekdays and weekends. The questionnaires were applied with the help of the ECCs supporters (e.g. socio-cultural technicians) and due to this fact the response rate was 100% for all studied sites.

Considering the time-budget data, an elderly daily pattern was achieved in order to identify the most occupied micro-environments. All the results were pondered to 24h.

#### 2.3.4 Air pollutants measurements in 10 ECCs

In a first stage, a monitoring programme was undertaken in ten ECCs where the IAQ was assessed during the occupied periods in two different indoor micro-environments: bedrooms and living-rooms. The bedrooms were chosen according to the occupancy – always two persons per bedroom. All the selected bedrooms were occupied by two elders to keep the occupancy as a constant and because this occupancy reflects the reality of the majority of the bedrooms in the studied ECCs. As the physical characteristics of all bedrooms in each ECC were equivalent it was decided to select only one bedroom per ECC and to perform longer measurements in order to identify temporal patterns. In each ECC, measurements in bedrooms were made during the night varying between 11 and 16 h, depending on ECC routine. All ECCs had one living-room except ECC 1 and ECC 2 that had two living-rooms with the same characteristics, and therefore only one of them was selected. Measurements in living-rooms were made during the day and varied between 7 and 13 h. The correspondent outdoor measurements were performed between 5 min and 16 h. Different methodologies were used according to 3 criteria: ECCs availability, elderly health status and equipment availability. The PM10 outdoor values for the first four ECCs were provided from gravimetric methods (which will be explained further in Chapter 2.3.5.1).

A set of pollutants were selected to characterize the IAQ inside those micro-environments, such as air temperature (T), relative humidity (RH), carbon dioxide (CO<sub>2</sub>), carbon monoxide (CO), particulate matter in 5 different sizes (PM0.3-0.5, PM0.5-1, PM1-2.5, PM2.5-5 and PM5-10), total volatile organic compounds (VOC), ozone (O<sub>3</sub>) and formaldehyde (CH<sub>2</sub>O). For each ECC an evaluation of outdoor pollutants was also performed. The sampling was repeated during three consecutive days and occurred between October to December of 2012, avoiding extreme temperatures and humidity. Table 2.3 summarizes the parameters analysed both indoors and outdoors and the used equipment. All instruments were calibrated by certified entities, where they calibrate, validate and demonstrate that the instrument it was suitable for its intended purpose.

Table 2.3 – Parameters assessed in the ECCs and used equipments.

Indoor						
Pollutant	CO <sub>2</sub> , CO, VOCt, O <sub>3</sub>	PM0.3-0.5, PM0.5-1, PM1-2.5, PM2.5-5 and PM5-10		CH <sub>2</sub> O		
	GrayWolf <sup>®</sup>	Light	house®	Si®		
Equipment	Equipment Direct Sense Handheld 3016		neld 3016	Formaldemeter htV-M		
Method	Photoionization probe	Diffusion optical light		Electrochemical formaldehyde sensor comprising two noble metal electrodes and a suitable electrolyte		
		Outdoo	or			
Pollutant	utant CO <sub>2</sub> , CO, VOCt, O <sub>3</sub> , T, RH			PM10		
Equipment	TSI® 754	5	TCR-Tecora			
Method	Sensors ty  NDIR; Electro-c Thermistor and capacitiv	hemical; Thin-film	Gravimetric: low volume with PM10 impactor using teflon filters			
Equipment	TSI® 754  Sensors ty  NDIR; Electro-c Thermistor and	5 pe: hemical; Thin-film	Gravimetric	TCR-Tecora		

#### 2.3.5 Air pollutants measurements in 4 ECCs

In a second stage, and after the previous analysis, the first four ECCs (ECC 1 to ECC 4) were selected in order to perform a deeper IAQ assessment. This selection was made according to different criteria: ECCs availability, geographical localization and ECCs characteristics. In these ECCs 1) the fungi contamination was evaluated; 2) the nanoparticles deposition in elderly lungs was estimated; and 3) the daily exposure and inhaled dose of institutionalized elders to PM10 components (trace elements and carbonaceous compounds) were calculated.

The bedrooms were chosen according to the occupancy – always two persons per bedroom. All the selected bedrooms were occupied by two elders to keep the occupancy as a constant and because this occupancy reflects the reality of the majority of the bedrooms in the studied ECCs. As the physical characteristics of all bedrooms in each ECC were equivalent it was decided to select only one bedroom per ECC and to perform longer measurements in order to identify temporal patterns. In each ECC, measurements in bedrooms were made during the night varying between 11 and 16 h, depending on ECC routine. All ECCs had

one living-room except ECC 1 and ECC 2 that had two living-rooms with the same characteristics, and therefore only one of them was selected. Measurements in living-rooms were made during the day and varied between 7 and 13 h (Almeida-Silva et al., 2014b).

The sampling was repeated during three consecutive days and took place between October to December of 2012, avoiding extreme temperatures and humidity.

#### 2.3.5.1 PM10 – sampling and determination of mass concentration

PM with an aerodynamic diameter lower than 10  $\mu$ m (PM10) was sampled in four ECCs during the occupied periods in the micro-environments selected as it was described previously. The sampling campaign occurred between October and November of 2012, avoiding extreme temperature and humidity.

In the indoor of each ECC, PM10 was collected with two TCR-Tecora® samplers operating at a flow rate of 2.3 m³.h⁻¹ (in accordance with the EN 12341, equipped with a PM10 EN sampling head). One sampler collected particles onto quartz filters and another onto teflon filters, both with a diameter of 47 mm. The sampling time ranged from 10 to 16 h, during the occupancy period for each selected micro-environment: bedroom and living-room. Outdoor measurements were performed in parallel with two different samplers: 1) a Partisol™ Plus 2025 Sequential Ambient Particulate Sampler, operating at a flow rate of 1.0 m³.h⁻¹ and collecting particles onto teflon filters and 2) a Leckel Medium Volume Sampler 6, operating at a flow rate of 3.5 m³.h⁻¹ and collecting particles onto quartz filters.

Carbonaceous components were determined in PM10 deposited onto quartz filters whereas teflon filters were used to measure the aerosol elemental composition. The collected filters were weighted using a Mettler Toledo balance with 0.1  $\mu$ g readability. The balance was placed in a controlled clean room (class 10,000) at a temperature of  $20\pm1^{\circ}$ C and a relative humidity of  $50\pm5\%$ . Before being weighted, filters were allowed to be equilibrated during 24 hours in the same room. Filters were weighted before and after sampling and the mass was obtained as the average of three measurements, when observed variations were less than 5  $\mu$ g.

In order to assure the comparability of the PM10 results provided from different devices and with different matrix an inter-comparison was performed between the four used gravimetric equipments with themselves and with diffusion optical light equipment. Figure 2.3 shows a good agreement between equipments and matrixes. The results showed that Lighthouse overestimate the levels in comparison to gravimetric method, as already been shown by Yanosky and co-workers (2002), but presents high correlations when compared to gravimetric samplers using different matrix ( $r^2 = 0.8$  and  $r^2 = 0.7$  for quartz and teflon, respectively). Fig. 2.4 also presents good correlations between quartz and teflon filters ( $r^2 = 0.9$  and  $r^2 = 0.8$  for outdoor and indoor, respectively).



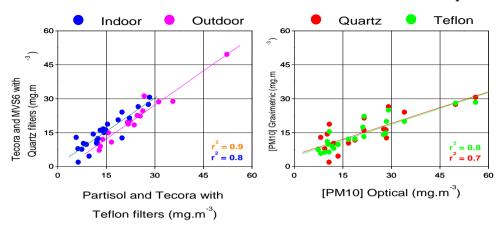


Figure 2.3 – Correlation between the four used gravimetric equipments with themselves and with diffusion optical light equipment.

#### 2.3.5.2 PM10 – carbonaceous components and trace elements

The elemental and organic carbon (EC and OC) content in PM10 was analysed in Aveiro University by a home-made thermal-optical transmission system, after a passive exposure of the sampled filters to vapours of hydrochloric acid (HCl - 6M) for approximately 4 hours to remove carbonate interferences. After this period, the samples were kept overnight in a desiccator with hydroxide sodium (NaOH) to neutralize any excess of acid in the sample. This procedure was at first developed by Carvalho et al. (2006) and recently adapted by Alves et al. (2011). Controlled heating in anoxic and oxic conditions was performed to separate, respectively, OC into two fractions of increasing volatility and EC, which were then measured in the form of CO<sub>2</sub> by an infrared non-dispersive analyser. The first fraction corresponded to the volatilization at T < 200°C of lower molecular weight organics. The second fraction was related to the decomposition and oxidation of higher molecular weight species at temperatures ranging from 200 to 600°C. However, pyrolyzed organic carbon (PC), formed during the previous heating steps, was only released in oxic conditions, when the sample was heated up to 850°C, evolving simultaneously with EC. The interference between PC and EC was controlled by continuous evaluation of the blackening of the filter using a laser beam and a photodetector that measured the light transmittance. The split between the PC and EC was assigned when the initial (baseline) value of the filter transmittance was reached. All carbon removed before the split was considered organic, and that removed after the split was considered elemental.

Carbonates  $(CO_3^{2-})$  in PM10 samples were analysed through the release of  $CO_2$ , and measured by the same non-dispersive infrared analyser coupled to the thermo-optical system, when a punch of each filter was acidified with orthophosphoric acid (20%) in a free  $CO_2$  gas stream (Alves, et al., 2011).

Trace elements in filters were measured by  $k_0$ -Instrumental Neutron Activation Analysis ( $k_0$ -INAA) in IST (Almeida et al, 2013a). One half of the filter was rolled up, put into an aluminium foil and irradiated at the Portuguese Research Reactor (nominal power: 1MW) during 5 h. After the irradiation, filters were removed from the aluminium foil and were inserted in polyethylene containers. Samples were measured during 7-10 hours after 2-5 days and 4 weeks of decay, in a coaxial germanium detector, associated to an ORTEC® Automatic Sample Changer. A comparator – Al-0.1% Au alloy disk with a thickness of 125  $\mu$ m and a diameter of 0.5 cm – was co-irradiated with the samples for the application of the  $k_0$ -INAA methodology (De Corte, 1987).

Quality control was pursued by the use of the NIST-SRM<sup>®</sup> 1633a – Coal Fly Ash certified reference material (Dung et al., 2010, Almeida et al, 2014b). Approximately 130 mg of reference material was co-irradiated with each batch of samples and measured for 1 h after 2-5 days and 4 weeks of decay using the same detector. Figure 2.4 shows a good agreement, with deviations from reference values below 20%, with 95% of confidence level (Almeida-Silva et al., 2014c).

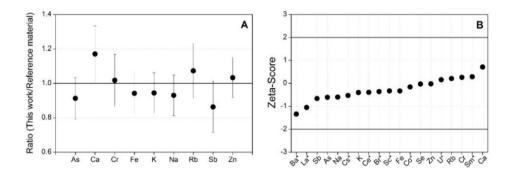


Figure 2.4 – A: Ratio between the results obtained in this work for NIST-SRM® 1633a "Coal Fly Ash" and certified values. Uncertainty is at 95% confidence level based on t-Student distribution. B: Plot of zeta-score calculated for NIST-SRM® 1633a "Coal Fly Ash": certified and consensual values. (\* Consensual values according to Roelandts and Gladney (1998)).

Three of each blank teflon and quartz filters were treated at the same conditions as samples. The carbonaceous components and the element concentrations were corrected according to blank results by subtracting those values.

#### 2.3.5.3 Nanoparticles – measurements and equipment

In this study a nanoparticle surface area monitor (NSAM) (TSI, Model 3550; Shoreview, MN) was used to measure the lung-deposited surface area of particles which is expressed as square micrometres of lung surface per cubic centimetre of inhaled air (µm².cm⁻³). Nanoparticles are described to have an increasing surface area with a decreasing particle size for the same amount of mass, being very desirable the determination of nanoparticle surface area deposited in the human lung. This deposition corresponds to the tracheobronchial or alveolar regions of the human lung, according to the International Commission on Radiological Protection (ICRP) deposition model developed by the American Conference of Governmental Industrial Hygienists (ACGIH) (ICRP, 1994).

This equipment is based on diffusion charging of sampled particles, followed by detection of the charged aerosol using an electrometer. An aerosol sample is drawn into the instrument continuously at a flow rate of 0.15 m<sup>3</sup>.h<sup>-1</sup>. The flow is split with 0.06 m<sup>3</sup>.h<sup>-1</sup> passing through two filters (a carbon and a HEPA) and an ionizer and 0.09 m<sup>3</sup>.h<sup>-1</sup> of aerosol sample flow. The flow streams are merged in a mixing chamber where particles in the aerosol flow mix with the ions carried by the filtered clean air. This patented counter-flow diffusion charging brings the aerosol particles into a defined, charged state. The separation of particles from direct interaction with the corona needle and/or the strong field near it reduces particle loss and makes the charging process more efficient and reproducible. The charged aerosol then passes through an ion trap to remove excess ions and charged aerosol. The aerosol then moves-on to an electrometer for charge measurement. In the electrometer, current is passed from the particles to a conductive filter and measured by a very sensitive amplifier, as shown schematically in Figure 2.5.

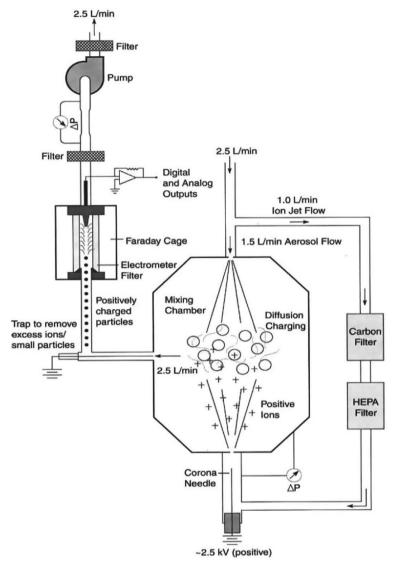


Figure 2.5 – Schematic showing the operation principle of a NSAM equipment (TSI, 2012).

The charge measured by the electrometer is directly proportional to the surface area of the particles passing through the electrometer. The equipment was set to alveolar response settings only, as this is the most significant metric.

#### 2.3.5.4 Fungi – samples collection, preparation and analysis

Air fungal contamination was studied by conventional and molecular method in 5 different micro-environments: bedrooms, living-rooms, canteens, storage and outdoors, at one meter

height. Surface fungal contamination was also assessed in all the same micro-environments, unless outdoor.

#### **Conventional methods**

Using conventional methods air samples were collected through an impaction method by a Microbiological Air Sampler (MAS-100TM) with an airflow rate of 8.4 m $^3$ .h $^{-1}$  onto malt extract agar (MEA) supplemented with the antibiotic chloramphenicol (0.05%). In the same indoor environments surfaces were swabbed using a 10 by 10 cm square stencil that was disinfected with 70% alcohol solution between samples according to the International Standard ISO 18593. The obtained swabs were then plated into MEA media. All collected samples were incubated at 27  $\pm$  2°C for 5 to 7 days. After lab processing and incubation of collected samples, quantitative and qualitative results were obtained with identification of the isolated fungal species.

In order to understand the possible influence of human occupancy on fungal contamination, an additional approach was applied. Fungal contamination was assessed before and after occupancy in two different indoor micro-environments: bedroom and living-room. The air samples were collected as described previously.

#### Molecular analysis

For molecular analysis, 20 air samples of 250 L were collected from the four ECCs using the Coriolis µ air sampler (Bertin Technologies), at 18 m<sup>3</sup>.h<sup>-1</sup> airflow rate. Each air sample was collected into a conic sterile tube containing 10 ml sterile phosphate-buffered saline and 0.05% Triton X-100. Five ml from the collection liquid were centrifuged at 2500 x g for 10 min and supernatant removed to leave a 250 µl pellet that was subsequently used for DNA extraction. DNA was then extracted using the ZR Fungal/Bacterial DNA MiniPrep Kit (Zymo Research) according to the manufacturer's recommendations. Molecular identification of Aspergillus fumigatus was achieved by a Real Time PCR (RT PCR) using the Rotor-Gene 6000 qPCR Detection System (Corbett) under specific cycling conditions and with specific primers and probes (Table 2.4), Reactions included 1× iO Supermix (Bio-Rad), 0.5 μM of each primer, and 0.375 μM of TaqMan probe in a total volume of 20 μl. Amplification followed a three-step PCR reaction: 40 cycles with denaturation at 95 °C for 30 seconds, annealing at 52 °C for 30 seconds, and extension at 72 °C for 30 seconds. The specificity of the primers and probe set was confirmed by testing these primers in DNA extracted from pure cultures of different species from the same genus (A. fumigatus, A. flavus, A. terreus, A. niger, A. versicolor, and A. ochraceus).

Table 2.4 – Specific primers and TaqMan probes used in real-time PCR for DNA amplification of isolates belonging to *Aspergillus fumigatus* complex.

			Reaction Conditions		
A. fumigatus	Sequence -	Concentration (µM)	Ann T ( <sup>a</sup> C)		
(Cruz-Perez et al., 2001) F: CGCGTCCGGTCCTCG					
, , , , ,	R: TTAGAAAAATAAAGTTGGGTGTCGG	0.375	52		
	P: FAM-TGTCACCTGCTCTGTAGGCCCG-TAMRA				

#### 2.3.6 Statistical analysis

Statistical analysis of all data was performed using the STATISTICA<sup>®</sup> software. The Mann–Whitney U-test and Wilcoxon Matched pairs were applied to detect statistically significant differences between each indoor micro-environment (independent samples) and between indoor and outdoor results (dependent samples), respectively. The criterion for significance was set at p < 0.05. All data were graphically represented using Origin<sup>®</sup> software version 7.5 (OriginLab).

#### 2.4 Results and Discussion

#### 2.4.1 Elderly daily pattern

Several studies have already evaluated the daily time pattern of people from different countries (Fisher and Robinson, 2011; Eurostat, 2006, 2003). However, these studies either excluded the old people or studied simultaneously all age groups, since young children to elderly.

Therefore, due to the scarcity of works focusing on time occupancy by elders living in ECCs, a questionnaire was applied to build a time budget survey for the studied population and the following conclusions were achieved: 1) the majority of the elders spend their time principally in bedrooms and living-rooms; 2) a few percentage of old people went outside to stay in the ECC' garden or to go to another indoor places as family houses, restaurants or coffees; and 3) 7% of the elders (22 women and 6 men) were bedridden and, therefore, they were always inside their bedrooms.

The micro-environment "others" corresponds to other indoor micro-environments rather than the living-room or bedroom. For the calculation of exposure and inhaled dose, equivalent characteristics as the living-rooms were used for this micro-environment.

Figure 2.6A shows the time spent by the elders in each micro-environment. Due to the lack of differences between weekdays and weekends the results were presented for a typical 24 h. Figure 2.6B indicates that elders living in the studied ECCs spent in average 95% of their time indoors. A study developed in Italy showed that elderly spent 70-83% of their time inside the buildings (Simoni et al., 2003). The National Human Activity Pattern Survey (NHAPS) developed in USA refers that the American elders spent 87% of their time indoors (Klepeis et al., 2001). These values were lower than the ones obtained in this work, which could be explained by the fact that the present study only considered elders living in ECCs. Old people in ECCs spent the majority of their time inside bedrooms (57%) and living-rooms (30%). In ECC 3 people spent more time in bedrooms due to the high number of bedridden (13%) in this institution. On the other hand, in ECC 1 and ECC 7 all the elders were daily lifted and placed in the living-rooms, which can explain the less percentage of time spent inside the bedrooms.

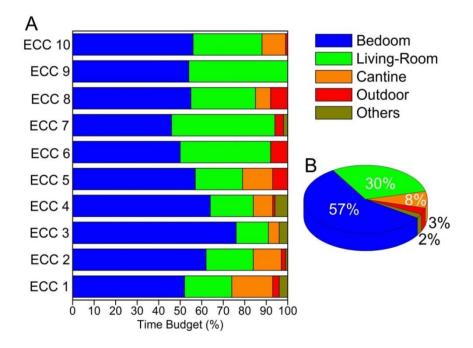


Figure 2.6 – A: Time budget-survey data for all 384 voluntaries (values in percentage); B: Time budget data per studied ECCs (values in number of hours).

#### 2.4.2 CO<sub>2</sub> concentrations, ventilation and air exchange rates in 10 ECCs

According to Dimitroulopoulou (2012) ventilation is recognized as an important component of a healthy dwelling, because it is possible to associate a poor/inadequate ventilation with indoor air pollution and, consequently, human health problems. Elderly

are considered a susceptible population and a group with more resistance to change old habits, which could hamper the promotion of ventilation and lead to an increase of exposure to indoor air pollution (Canha et al., 2013; Coelho et al., 2005). In Portuguese legislation (Portaria 353-A/2013) the minimum ventilation rates for bedrooms and livingrooms are 4.4 lps.person<sup>-1</sup> and 5.6 lps.person<sup>-1</sup>, respectively. Air exchange rates ( $\alpha$ ) – renovation per hour (h<sup>-1</sup>) – and ventilation rates (Q1) – air litters per second per person (lps.person<sup>-1</sup>) – were calculated for all sites using the build-up method based on CO<sub>2</sub> concentrations as a tracer gas (Canha et al., 2013; Hänninen, 2013). Using CO<sub>2</sub> as tracer gas represents an advantage comparing with other tracer since it is emitted by occupants and it is inert (Hänninen, 2013). Since this method is focused on school classrooms that shifts strongly with time, as the opposed to what happens in ECCs, the method was adapted. The build-up curves used in this work corresponded to the periods that recorded changes on occupancy, e.g. lunch time. Table 2.5 presents the air exchange rates, the ventilation rates and the CO<sub>2</sub> concentrations for the studied sites. Also shows that α ranged between 0.2 and 2 h<sup>-1</sup> for bedroom and between 0.7 and 2.0 h<sup>-1</sup> for living-room while O1 varied between 1.7 and 19 lps.person<sup>-1</sup> in bedroom and between 0.9 and 3.2 lps.person<sup>-1</sup> in living-room. Living-rooms were, in almost all ECCs, the micro-environment with lower O<sub>1</sub> values due to higher number of people that occupied these spaces, 30% and 100% of the total indoor micro-environments evaluated did not meet the Portuguese legislation for bedrooms and living-rooms, respectively. According to the main national standards in Europe (but despite the lack of unanimity), the air exchange rate of 0.5 h<sup>-1</sup> is defined as a threshold below which associations with poor IAQ may occur (Dimitroulopoulou, 2012). In this study only one bedroom (ECC 10) presented an air exchange rate lower than this value. Comparing with ASHRAE guidelines, 35% of the evaluated micro-environments met the established value of 5.5 lps.person<sup>-1</sup> (ASHRAE, 2004). ASHRAE does not define specific minimum ventilation rates for ECCs, but establishes the guideline of 8.6 lps.person<sup>-1</sup> for children care centers. Considering that both populations are susceptible, this guideline was compared with the values presented in Table 2.5 and it was verified that only 2 microenvironments (ECC 6 and ECC 9 bedrooms) met this requirement.

Table 2.5 – Air exchange rates ( $\alpha$ ) and ventilation rates (Q<sub>1</sub>) assessed in 10 ECCs, for both bedroom and living-room.

Micro-e	nvironments	α (h <sup>-1</sup> )	STD [α (h <sup>-1</sup> )]	Q <sub>1</sub> (lps.person <sup>-1</sup> )	STD [Q <sub>1</sub> (lps.person <sup>-1</sup> )]	CO <sub>2</sub> [avg (STD)] (mg.m <sup>-3</sup> )
ECC 1	Bedroom	1.1	0.8	8.3	5.0	1600 (450)
	Living-Room	0.8	2.7	3.2	4.5	1050 (190)
ECC 2	Bedroom	1.1	0.2	6.8	1.1	1400 (270)
	Living-Room	1.5	1.0	2.0	1.2	1200 (220)
ECC 3	Bedroom	0.7	1.4	8.2	9.0	3000 (780)
2000	Living-Room	8.0	1.3	2.2	2.2	1700 (400)
ECC 4	Bedroom	0.50	0.07	3.0	0.41	2100 (960)
200 1	Living-Room	1.20	0.78	2.6	1.63	1700 (500)
ECC 5	Bedroom	1.60	0.89	1.7	1.23	3000 (570)
2000	Living-Room	1.60	0.89	2.5	1.9	1200 (390)
ECC 6	Bedroom	1.70	0.88	16.3	7.3	2400 (260)
2000	Living-Room	0.70	0.46	3.0	1.6	2200 (620)
ECC 7	Bedroom	1.8	0.8	8.1	5.0	2300 (890)
2007	Living-Room	2.0	1.0	0.9	0.46	570 (110)
ECC 8	Bedroom	2.0	0.1	7.0	0.39	890 (70)
2000	Living-Room	1.3	1.1	1.4	0.98	810 (200)
ECC 9	Bedroom	1.4	0.7	19.3	8.6	680 (120)
200 9	Living-Room	1.6	0.8	4.7	3.0	660 (120)
ECC 10	Bedroom	0.20	0.15	2.8	1.9	1900 (320)
	Living-Room	1.3	1.9	3.2	2.7	1700 (400)

 ${\rm CO_2}$  is a pollutant emitted by human metabolism and is commonly used as an indicator of occupancy and poor ventilation (Almeida et al., 2011).  ${\rm CO_2}$  concentrations in bedroom were significantly higher than in living-room (p < 0.05). In 40% of the studied bedrooms the  ${\rm CO_2}$  limit value (2250 mg.m<sup>-3</sup>) defined by the Portuguese legislation was exceeded.  ${\rm CO_2}$  concentration exceeded the Portuguese legislation in 84% of the measuring time (15 h) and presented an average value of 3160 mg.m<sup>-3</sup> (Portaria 353-A/2013).  ${\rm CO_2}$  presented a

similar trend in all ECCs bedrooms. As an example, Figure 2.7 presents the temporal variation of  $CO_2$  concentration evaluated in ECC 5 during the first sampled night. It was possible to observe a huge increase since the evening until the elderly uprising. In fact, ECC 5' bedroom was the indoor micro-environment that presented significantly highest  $CO_2$  concentrations comparing with all the other studied micro-environments. The exception was the bedroom of ECC 3 which did not present significantly differences comparing with bedroom of ECC 5 (p = 1.0) and was the second site with the highest  $CO_2$  concentrations.

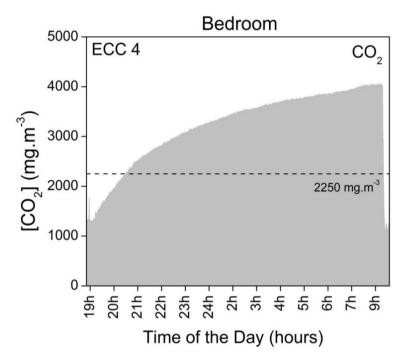


Figure 2.7 – Temporal variation (time of the day in hours) of  $CO_2$  in ECC 4 bedroom (values in mg.m<sup>-3</sup>).

# 2.4.3 CO, $O_3$ , VOC, $CH_2O$ , PMx concentrations and comfort parameters in 10 ECCs

Air temperature, relative humidity, carbon monoxide, particulate matter in 5 different sizes, total volatile organic compounds, ozone and formaldehyde were measured in order to perform a complete IAQ assessment. Table 2.6 summarizes the results of all measured parameters, in order to compare each site and pollutants. Results showed that in average PM concentrations in living-rooms were significantly higher than in bedrooms, except for ECC 7 (p = 0.79). Living-rooms of ECC 4, ECC 5, ECC 7 and ECC 10 presented the

highest PM10 average concentration (44, 43, 41 and 47  $\mu$ g.m<sup>-3</sup>, respectively). For ECC 4 and ECC 5, PM10 maximum values were 860 and 350  $\mu$ g.m<sup>-3</sup>, respectively. The high PM10 concentration measured in ECC 7 can be explained by the high outdoor PM10 concentration (71  $\mu$ g.m<sup>-3</sup>). The bedrooms of ECC 7 and ECC 10 presented a PM2.5 average concentration of 21  $\mu$ g.m<sup>-3</sup> and 34  $\mu$ g.m<sup>-3</sup>, respectively. In order to identify the possible sources of particles, Figure 2.8 shows the temporal variation of particles in different microenvironments. In bedroom graphs it is possible to observe the particles deposition during the night and then its re-suspension while elderly uprising. The observed peaks of particles assessed in ECC 10 during the night were due to the control done by the supporters. Considering the living-room, two different particles' behaviours were observed: in ECC 4 elders went to the living-room at 8 AM promoting the re-suspension of particles and consequently the increasing of its concentrations; and in ECC 5 a high peak of particles was observed during the mid-day which is explained by the fact that in this ECC meals were served in the living-room.

The average particle concentrations measured in this work were similar to those found in a study developed with elders living in Amsterdam and Helsinki that presented PM2.5 average concentrations of 16 µg.m<sup>-3</sup> and 11 µg.m<sup>-3</sup>, respectively (Lanki et al., 2007). In UK houses PM10, PM2.5 and PM1 concentrations (13, 6 and 3 µg.m<sup>-3</sup>, respectively) were lower comparing with current work (Nasir and Colbeck, 2013). PM2.5 and PM10 average concentrations measured in several houses demonstrated a similarity of results: 8 µg.m<sup>-3</sup> and 17 μg m<sup>-3</sup> (Jones et al., 2000); 9 μg.m<sup>-3</sup> and 23 μg.m<sup>-3</sup> (Lawson et al., 2011); 7 μg.m<sup>-3</sup> and 22 µg.m<sup>-3</sup> (Molloy et al., 2012). However, it is possible to find studies with higher particles concentrations, such as the one developed by Chao and Wong (2002) where PM2.5 and PM10 average concentration of 45 µg.m<sup>-3</sup> and 63 µg.m<sup>-3</sup> were measured, respectively. Those values could be explained by the existence of different sources: smoking, cooking and burning incense (Chao and Wong, 2002). Comparing the livingroom with the bedroom it is possible to observe that the coarse fraction is dominant in living-room whereas fine fraction is dominant in bedroom. This fact indicates the importance of re-suspension in living-room. As it was said before, elderly are considering a susceptible population such as children. By this reason and because studies performed in ECCs are rare, results from the current work were also compared with the ones developed in schools. The largest difference between ECCs and schools is the behaviour of the population – most of the elders have reduced movement capacities whereas children are in constant activity promoting a higher re-suspension of dust. Studies developed in Lisbon schools showed that children were exposed to a PM2.5 concentration of 10 µg.m<sup>-3</sup> and the PM10 concentrations varied from 30 µg.m<sup>-3</sup> to 146 µg.m<sup>-3</sup> (Canha et al., 2012b; Almeida et al., 2011). Another study developed in three different Polish schools referred that average particles concentrations varied between 94 µg m<sup>-3</sup> and 191 µg.m<sup>-3</sup> for PM10 and 45 µg.m<sup>-3</sup> and 119 ug.m<sup>-3</sup> for PM2.5 (Polidnik, 2013).

Table 2.6 – Pollutants concentration assessed in 10 ECCs. PM10, PM2.5 and PM1 concentrations are presented in  $\mu g.m^{-3}$ , relative humidity in %, temperature in °C and the others concentrations are present in  $mg.m^{-3}$  (AVG= Average and STD= Standard Deviation).

EEC	Site	Sampled					Parameters [AVG (STD)]								
		time (h)	PM10	PM2.5	PM1	co	voc	CH <sub>2</sub> O	O <sub>3</sub>	RH	Temp				
	BR	36ª	11 (4)	4.6 (1.3)	3.1 (1.2)	0.55 (0.47)	1.1 (0.5)	0.070	0.010 (0.011)	60 (3)	25 (1)				
EEC1	Outdoor	36 a	26 <sup>b</sup>	-	-	0.01 (0.04)	-	-	-	74 (15)	19 (4)				
	LR	36 a	15 (6)	4.0 (0.2)	1.3 (0.3)	0.11 (0.31)	0.52 (0.36)	0.06	0.011 (0.011)	53 (4)	25 (1)				
	Outdoor	36 a	22 <sup>b</sup>	-	-	0.02 (0.04)	-	-	-	72 (10)	21 (3)				
	BR	36 a	12 (9)	3.7 (1.9)	1.8 (0.9)	0.06 (0.10)	0.10 (0.060)	-	-	56 (5)	23 (1)				
EEG2	Outdoor	36 ª	12 <sup>b</sup>	-	-	0.17 (0.15)	-	-	-	66 (11)	15 (4)				
EEC2	LR	36 ª	30 (9)	6.7 (2.9)	3.5 (1.8)	0.12 (0.36)	0.24 (0.32)	0.02	-	44 (6)	23 (1)				
	Outdoor	36 a	24 <sup>b</sup>	-	-	< 0.0°	-	-	-	69 (1.5)	16 (1.4)				
	BR	36 a	13 (9)	3.7 (1.3)	2.1 (0.6)	0.72 (0.91)	0.37 (0.58)	-	-	52 (6)	23.0 (0.4)				
	Outdoor	36 a	18 <sup>b</sup>	-	-	0.13 (0.040)	-	-	-	63 (9)	13 (2)				
EEC3	LR	36 a	28 (11)	9.4 (3.6)	6.0 (2.7)	0.26 (0.26)	0.23 (0.19)	-	-	52 (3)	24 (1)				
	Outdoor	36 a	24 <sup>b</sup>	-	-	0.17 (0.18)	-	-	-	70 (7)	16 (3)				
	BR	36 a	17 (15)	6.9 (4.9)	4.4 (3.2)	0.39 (0.31)	0.24 (0.12)	-	-	55 (4)	19.0 (0.4)				
	Outdoor	36 a	31 <sup>b</sup>	-	-	0.36 (0.31)	-	-	-	63 (5)	11 (3)				
EEC4	LR	36 a	44 (47)	10 (3.1)	4.4 (1.5)	0.35 (0.69)	0.39 (0.34)	-	-	63 (5)	21 (1)				
	Outdoor	36 a	17 <sup>b</sup>	-	-	0.13 (0.11)	-	-	-	76 (8)	17 (3)				
	BR	16	29 (35)	9.2 (5.2)	4.7 (2.6)	1.3 (1.6)	4.1 (1.5)	0.15	0.05 (0.19)	74 (1)	18 (1)				
EECs	Outdoor	-	-	-	-	-	-	-	-	-	-				
EEC5	LR	8	43 (61)	11 (22)	4.6 (8.3)	0.9 (1.4)	2.0 (0.3)	0.12	0.003 (0.010)	54 (7)	19 (1)				
	Outdoor	-	-	-	-	-	_	_	-	-	_				

a) Sampling was performed during 3 consecutive occupied periods. b) Results providing from gravimetric method.

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c) Below the detection limit.

Table 2.6 – Pollutants concentration assessed in 10 ECCs. PM10, PM2.5 and PM1 are presented in  $\mu g.m^{-3}$ , relative humidity in %, temperature in  ${}^{\circ}C$  and the others are present in mg.m $^{-3}$  (AVG= Average and STD= Standard Deviation) (continued).

EEC	Site	Sampled time (h)					Paramete [AVG (ST				
		time (n)	PM10	PM2.5	PM1	co	voc	CH <sub>2</sub> O	O <sub>3</sub>	RH	Temp
EEC6	BR	13	6.2 (2.4)	2.8 (0.6)	1.7 (0.5)	0.78 (0.17)	1.3 (0.1)	0.030	0.0097 (0.0017)	49 (3)	23 (1)
	Outdoor	-	-	-	-	-	-	-	-	-	-
	LR	10	27 (15)	11 (3.8)	7.5 (3.5)	0.13 (0.15)	1.20 (0.34)	-	0.02 (0.01)	46 (2)	23 (1)
	Outdoor	-	-	-	-	-	-	-	-	-	-
	BR	11	41 (21)	21 (12)	14 (10)	1.02 (0.54)	0.13 (0.21)	0.030	0.0029 (0.0046)	75 (4)	16.0 (0.2)
EEC7	Outdoor	0.1	71 (31)	23 (2)	4 (0.4)	-	-	-	-	-	-
EEC7	LR	9	35 (4.9)	16 (3.2)	5.1 (2.3)	0.10 (0.11)	$< 0.0^{\rm c}$	0.030	0.010 (0.002)	74 (2)	18 (1)
	Outdoor	0.1	71 (32)	23 (2.3)	4.4 (0.44)	0.04 (0.08)	$< 0.0^{\rm c}$	-	0.03 (0.01)	78 (3)	15 (1)
	BR	12	16 (3.9)	8.9 (1.4)	2.7 (0.50)	0.84 (0.47)	0.59 (0.61)	0.030	0.015 (0.005)	66 (3)	20 (1)
EEC8	Outdoor	0.2	58 (21)	20 (5.7)	3.6 (0.65)	0.10 (0.20)	< 0.0°	-	0.04 (0.04)	80 (3)	13.0 (0.3)
EEC8	LR	7	19 (7.2)	9.4 (4.6)	2.3 (0.8)	0.15 (0.13)	0.12 (0.10)	0.050	0.00002 (0.00050)	63 (7)	22 (1)
	Outdoor	0.2	45 (18)	14 (4.2)	3.1 (0.81)	0.36 (0.55)	0.15 (0.10)	-	0.03 (0.02)	78 (12)	14 (2)
	BR	13	11 (7.3)	4.7 (1.2)	2.7 (0.77)	0.033 (0.087)	0.36 (0.06)	0.030	0.0009 (0.0025)	53 (2)	16 (1)
EECO	Outdoor	0.2	18 (12)	6.4 (2.1)	3.7 (1.2)	0.18 (0.44)	0.21 (0.08)	-	0.04 (0.05)	48 (5)	9 (1)
EEC9	LR	10	11 (3.4)	4.2 (0.65)	2.7 (0.37)	0.0° (0.0)	0.41 (0.05)	0.030	0.040 (0.005)	53 (2)	16 (1)
	Outdoor	0.2	18 (12)	6.4 (2.1)	3.7 (1.2)	0.00030 (0.1.0)	0.31 (0.05)	-	0.04 (0.05)	48 (6)	9 (1)
	BR	14	47 (21)	34 (12)	28 (11)	1.9 (0.68)	0.0004 (0.0021)	0.030	0.002 (0.007)	75 (2)	18.0 (0.3)
	Outdoor	0.2	30 (15)	14 (3.9)	6.1 (3.1)	0.20 (0.30)	0.12 (0.14)	-	0.02 (0.02)	75 (6)	14 (1)
EEC10	LR	12	31 (3.0)	12 (1.0)	4.0 (0.10)	2.9 (1.6)	0.73 (0.82)	0.040	0.01 (0.24)	71 (2)	21 (1)
	Outdoor	0.2	43 (11)	26 (3.3)	12 (3.1)	0.80 (1.1)	0.05 (0.10)	-	0.02 (0.01)	83 (5)	14 (1)

a) Sampling was performed during 3 consecutive occupied periods. b) Results providing from gravimetric method.

c) Below the detection limit.

CO is essentially associated with combustion (Raub et al., 2000; Oliver et al., 1999) and the results showed that its concentration was significantly higher in bedrooms than in living-rooms (p = 0.00). CO concentrations measured in bedroom (1.9 mg.m<sup>-3</sup>) and in living-room (2.9 mg.m<sup>-3</sup>) of ECC 10 was significantly higher comparing with all the other evaluated indoor micro-environments (p = 0.00). The limit value defined by the Portuguese legislation (10 mg.m<sup>-3</sup>) was not exceeded (Portaria 353-A/2013). These results were higher when compared with a study developed in Delhi that found very low concentrations of CO in 9 bedrooms (Prasad et al., 2003).

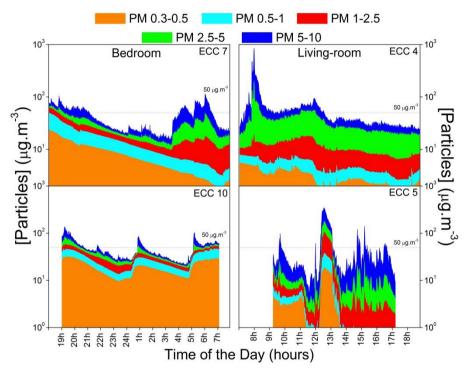


Figure 2.8 – PM0.3-0.5, PM0.5-1, PM1-2.5, PM2.5-5 and PM5-10 temporal variation (time of the day in hours) in ECC 7 and ECC 10's bedrooms and in ECC 4 and ECC 5's livingrooms (values in μg.m<sup>-3</sup>).

Ozone concentration was low in all ECCs, except for ECC 5 bedroom (maximum value of  $1.3 \text{ mg m}^{-3}$ ) and for ECC 1 living-room (maximum value of  $1.5 \text{ mg m}^{-3}$ ). As it is possible to observe in Figure 2.9 the peak of  $O_3$  concentration is presented in the bedroom during the elderly uprising. In living-room of ECC 1 the high peak of  $O_3$  concentration was observed during all morning. In fact, during the other periods of the day the  $O_3$  concentrations were too low or, in some cases, below the detection limit.

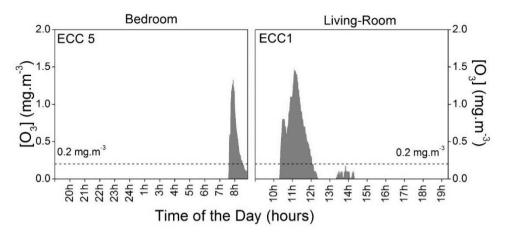


Figure 2.9 – Temporal variation (time of the day in hours) of  $O_3$  in bedroom of ECC 5 and living-room of ECC 1 (values in mg.m<sup>-3</sup>).

Formaldehyde and VOC are usually associated with indoor sources (Jantunen, 2007) that can be linked with furniture, cabinetry, carpet title, vinyl wall coverings, paints, adhesives, glue, varnish and cleaning products (Weschler, 2009; Valuntaite and Girgdiene, 2008; Bernstein et al., 2008). The Portuguese limit value for VOC (0.6 mg.m<sup>-3</sup>) was exceeded in 15 of the 20 analyzed indoor micro-environments, being the ECC 5 the one that presented the highest average concentrations – 4.1 and 2.0 mg.m<sup>-3</sup>, in bedroom and living-room, respectively. In order to identify the VOC sources, Fig 2.10 shows its temporal variation. In both cases VOC concentration was higher than the Portuguese guideline. In bedroom it was possible to observe a high peak in the morning period which is explained by the use of cleaning products during the elders uprising. In living-room besides the high VOC levels, no specific peaks were observed. The Portuguese limit value of 0.1 mg.m<sup>-3</sup> was not exceeded in the studied ECCs. Formaldehyde concentration was high in five microenvironments, being ECC 1 the one that presented the highest values – 0.07 mg.m<sup>-3</sup> and 0.06 mg.m<sup>-3</sup> – for bedroom and living-room, respectively.

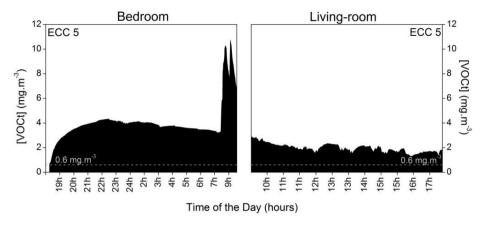


Figure 2.10 – Temporal variation (time of the day in hours) of VOC in ECC 5 bedroom and living-room (values in mg.m<sup>-3</sup>).

Regarding the comfort parameters, it is possible to affirm that the greater problem was related to the humidity. ECC 2, ECC 5, ECC 6, ECC 7 and ECC 10 presented relative humidity that come out of the range defined by international guidelines (50-70%) (ISO 7730). ECC 2 and ECC 6 presented results below the range (56% and 49% in bedroom and 44% and 46% in living-room, respectively). The other three presented values above the range (74%, 75% and 75% in bedroom, respectively, and 74% and 71% for living-room of ECC 7 and ECC 10, respectively).

# 2.4.4 PM10 and its components concentrations in 4 ECCs

Considering the fact that elders living in ECCs spent most of their time in bedrooms and living-rooms, PM10 was evaluated in these two indoor micro-environments. Figure 2.11 summarizes the indoor and respective outdoor PM10 concentrations measured in the 4 studied ECCs. The average PM10 concentration in bedroom and living-room was 11 µg.m<sup>-3</sup> and 19 ug.m<sup>-3</sup>, respectively. Living-rooms presented significantly higher PM10 concentrations when compared with bedrooms (p = 0.006) because living-rooms had more occupants. Several studies had already showed a relation between high levels of occupancy and high PM10 concentrations due to the effect of the re-suspension of dust (Canha et al., 2014a). PM10 concentrations in bedroom were significantly lower comparing with the correspondent outdoor (p = 0.005) whereas no significantly differences were observed between living-rooms and the outdoor (p = 0.590). These results are probably explained by the fact that during the occupancy period bedrooms had the windows closed while in living-rooms the doors and windows were open promoting a bigger circulation of air from the outdoor. PM10 indoor concentrations neither exceed the Portuguese guideline (50 μg.m<sup>-3</sup>) for indoor air quality (Portaria 353-A/2013) nor the reference value of 50 μg.m<sup>-3</sup> established by the World Health Organization (2010). A study developed in UK residences

presented similar results with PM10 average concentrations measured in living-rooms ranging between 13 ug.m<sup>-3</sup> and 22 ug.m<sup>-3</sup> (Nasir et al., 2013). However, comparing the current results with the majority of studies developed in different public indoor microenvironments it is possible to observe that PM10 concentration evaluated inside these ECCs were lower. Indeed, in these indoor micro-environments the movement of people is limited since most of the institutionalized elders were semi-autonomous. Consequently, the possibility of re-suspension of dust is lower comparing with other crowded indoor environments such as hospitals, schools and offices (Slexakova, et al., 2012; Canha, et al., 2012b). For instance, a study developed in a Portuguese hospital showed PM10 variations between 13 µg.m<sup>-3</sup> and 59 µg.m<sup>-3</sup> (Slexakova, et al, 2012). Zwozdziak and her co-workers (2013) studied the indoor air quality in schools from Poland and showed a range of PM10 concentrations between 43 µg.m<sup>-3</sup> and 69 µg.m<sup>-3</sup> (Zwozdziak, et al., 2013). Another study performed in Portuguese schools presented much higher PM10 concentrations, which varied between 30 µg.m<sup>-3</sup> and 146 µg.m<sup>-3</sup> (Almeida et al., 2011). In offices, several works also presented higher PM10 concentrations than the concentration assessed in current work, with a variation from 10 µg.m<sup>-3</sup> to 480 µg.m<sup>-3</sup> (Han, et al., 2011; Valuntaité, et al., 2008; Reynolds, et al., 2001).

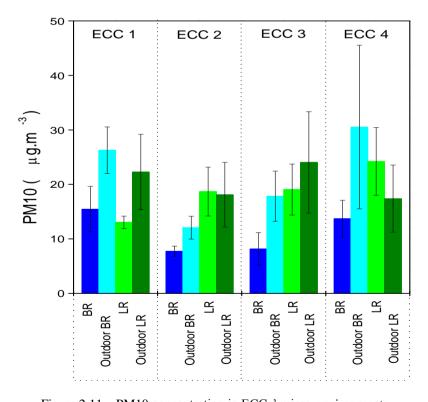


Figure 2.11 – PM10 concentration in ECCs' micro-environments.

Besides the low concentrations measured in this study it is well-known that PM10 enhances adverse health effects and it is unclear whether a threshold concentration exists for PM below which no effects on health are likely.

Table 2.7 summarizes the average and the standard deviation of the PM10 components. On average, the sum of the indoor PM10 components (OC, EC,  ${\rm CO_3}^{2-}$  and trace elements) measured in this work corresponded to 51% of the total PM10 mass measured by gravimetry.

The major component measured in PM10 was the carbonaceous fraction (OC, EC and  ${\rm CO_3}^{2-}$ ), representing in average 47% of the total indoor PM10 mass measured by gravimetry. Results showed that the concentration of these compounds were significantly higher indoors (p < 0.05). On average, OC accounted for 28% of the mass of PM10 indoors, whereas a lower mass fraction was found outdoors (10%), which represented an I/O ratio of 2.7. The I/O ratio for EC and  ${\rm CO_3}^{2-}$  presented lower values due to the influence of traffic and dust re-suspension at roadside, respectively.

The lack of correlation between indoor and outdoor EC and, principally OC, could be due to significant contributions of indoor sources. Cleary, OC was enriched in indoor as compared to outdoor. Among the indoor sources of organic compounds, sub-micrometer fragments of paper, skin debris, clothing fibers, cleaning products and waxes may be considered (Alves et al., 2014).

Organic to elemental carbon (OC/EC) ratios exceeding 2.0 have been used to identify the presence of secondary organic aerosols (SOA) (Chow et al., 1996) in urban areas where traffic emissions have a significant contribution to the EC levels. In this study, the average indoor OC/EC ratio was 5.4, which denotes a significant increase in OC levels that could be not only to primary indoor sources but as well to SOA production. The formation of SOA in indoor environments was demonstrated and confirmed in test chambers experiments, done by Aoki and Tanabe (2007). Sportion of semi-volatile organic compounds could be, as well as, important indoor source of organic matter. Thus, the combination of active indoor sources, sorptive processes and SOA formation leaded to an enrichment of the indoor particles in OC and to high OC/EC ratios inside the ECCs (Alves et al., 2014). Besides the lowest concentrations of OC and EC measured in bedrooms, the ratio OC/EC was higher in bedrooms (7.7) than in living-rooms (3.3), not only due to the higher contribution of skin debris, clothing fibers and cleaning products in bedrooms, which had a smaller area, but also due to less exchange with outdoor air, increasing the probability of SOA formation.

All the measured PM10 trace elements presented significantly higher levels in living-rooms than in bedrooms, except Ce that did not present significant differences between both micro-environments. Living-rooms had more occupancy, thus indoor concentrations of dust particles were strongly influenced by activities and movement of occupants, which may allow the re-suspension of previously deposited particles or their delayed deposition or settling. Moreover, the infiltration of outdoor air without any filtration was higher in living-

rooms, increasing the contribution of outdoor sources into this micro-environment (Almeida-Silva et al., 2014a).

Table 2.7 – Summary of bedrooms and living-rooms constituent concentrations and the respective outdoors.

	Bedroom	Outdoor (Bedroom)	Living-room	Outdoor (Living-room)
		[AVG	(STD)]	
PM10 (μg.m <sup>-3</sup> )	11 (4.4)	24 (12)	19 (6)	21 (8)
Carbonaceous compo	onents (µg.m <sup>-3</sup> )			
OC	3.0 (1.2)	2.2 (0.9)	4.3 (1.5)	1.7 (0.8)
EC	0.69 (0.45)	0.67 (0.42)	1.45 (0.66)	1.2 (1.0)
$CO_3^{2-}$	1.3 (0.5)	1.5 (1.5)	2.0 (1.0)	1.6 (1.2)
Elements (ng.m <sup>-3</sup> )				
As	0.28 (0.36)	0.9 (1.5)	0.33 (0.22)	0.94 (0.65)
Ce	0.47 (0.37)	0.35 (0.10)	0.37 (0.14)	0.42 (0.07)
Co	0.063 (0.050)	0.19 (0.15)	0.11 (0.06)	0.21 (0.07)
Cr	9.2 (4.4)	9.1 (5.3)	12(2)	14 (6)
Fe	48 (29)	270 (220)	225 (190)	450 (390)
K	100 (52)	960 (1460)	160 (80)	800 (960)
Na	201 (171)	920 (460)	435 (120)	1850 (1630)
Sb	0.49 (0.34)	1.9 (1.3)	1.4 (1.2)	2.2 (1.9)
Sc	0.0040 (0.0031)	0.025 (0.019)	0.018 (0.017)	0.033 (0.027)
Sm	0.0029 (0.0010)	0.017 (0.011)	0.013 (0.009)	0.047 (0.047)
Zn	26 (14)	27 (23)	39 (16)	32 (19)

Figure 2.12 shows that only Zn and Cr in some ECCs presented I/O ratios higher than 1, indicating the existence of indoor sources for these elements, although these elements are normally associated with traffic and industrial emissions (Zechmeister et al., 2005). Actually, other studies, which had also observed significantly higher concentration of Zn and Cr indoors (Chithra and Nagendra, 2013), suggested that Zn provides from several products which are applied indoors to protect steel, walls, wood surfaces, doors and windows (Avigo et al., 2008) and Cr can be derived from burning coal and kerosene (Joshi et al., 2010). The elements that presented higher concentration outdoors are essentially associated with outdoor anthropogenic sources related to traffic and industry (As, K and Sb) (Almeida et al., 2009), dust re-suspension (Ce, Co, Fe, K, Sc and Sm) (Almeida-Silva et al., 2011; Almeida et al., 2008), biomass burning (K) (Canha et al., 2012c) and sea spray (Na) (Almeida et al., 2013b). The higher or less decrease of I/O ratios observed among the different elements can also find an explanation in the size distribution associated with each element. Coarse particles fall down faster than fine particles, decreasing the particles population in the highest sizes. Elements with the lowest I/O ratios probably are associated to the coarse aerosol fraction (2.5 - 10 μm), whereas an I/O ratio close to one can be mainly associated to fine particles ( $dp < 2.5 \mu m$ ).

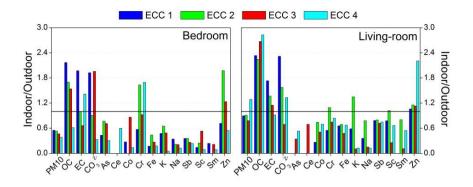


Figure 2.12 – Ratio indoor/outdoor for PM10 and its components

## 2.4.5 Fungal assessment in 4 ECCs

#### Fungal assessment by conventional methods

Figure 2.13 shows the total fungal load for all micro-environments assessed in the 4 ECC. The results ranged from 32 colony forming unit (CFU.m<sup>-3</sup>) in the bedroom of ECC 1 to 228 CFU.m<sup>-3</sup> in the storage room of ECC 4. On average, the living-room and storage area were the two micro-environments with lowest and highest fungal load, 58 CFU.m<sup>-3</sup> and 118 CFU.m<sup>-3</sup>, respectively. This may be attributed to the fact that all living-rooms had a cleaning frequency of once per day while the storage room was full of nutrients that favour fungal growth (Ekhaise et al., 2008). ECC 1 and ECC 3 presented the higher outdoor fungal load comparing with indoor micro-environments, 88 CFU.m<sup>-3</sup> and 92 CFU.m<sup>-3</sup>, respectively. Thus, data suggest an influence of outdoor air on IAQ related to fungal penetration (Goyer et al., 2001). Both ECC were located in a sub-urban area with a large green area in the surroundings. On the other hand, in all indoor micro-environments of ECC 2 the ratio indoor/outdoor (I/O) was higher than 1. Storage of ECC 4 was the other micro-environment with the ratio I/O above 1 suggesting a fungal indoor contamination (Nevalainen, 2007; Rao et al., 1996).

None of the micro-environments assessed exceeded the reference value defined by Portuguese law of 500 CFU.m<sup>-3</sup> (Portaria 353-A/2013). The storage area of ECC 1, where 228 CFU.m<sup>-3</sup> were isolated, revealed the highest fungal load. However, the occupants susceptibility are not consider in national legislation, since it is applied to several types of establishments such as schools, offices, hospitals, among others. Considering this fact, the results were also compared with a more demanding hospital threshold and it was observed that 6.3% of indoor micro-environments assessed exceeded the threshold defined by Krzysztofik in 1992 of 200 CFU.m<sup>-3</sup> (Augustowska & Dutkiewicz 2006).

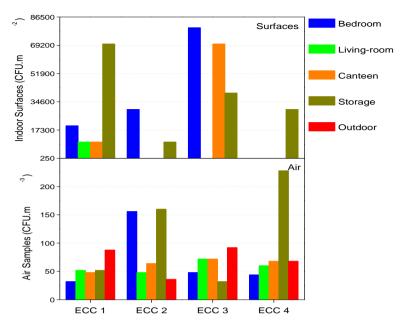


Figure 2.13 – Air and surfaces fungal load in four ECCs (values in CFU.m<sup>-3</sup> and CFU.m<sup>-2</sup>, respectively).

Twenty one different fungal species in indoor air were detected in a total of 1573 isolates. Table 2.8 also shows that *Penicillium* sp. was the most frequently isolated species (38%), followed by *Aspergillus* sp. (16%), and *Chrysonilia* sp. (4%). These results were similar to those obtained in a study conducted in children care centers (Zuraimi et al., 2009). Besides these fungal genera, other fungi were also identified: *Acremonium* sp., *Cladosporium* sp., *Neoscytalidium* sp., *Chrysosporium* sp., *Geotrichum* sp., *Alternaria* sp., *Scopulariopsis* sp., *Beauveria* sp., *Ulocladium* sp., *Aureobasidium* sp. and *Paecilomyces* sp. Other studies assessing fungal contamination also found *Penicillium* sp., *Aspergillus* sp., *Cladosporium* sp. and *Alternaria* sp. as the most prevalent fungi detected (Zhen-Feng et al., 2011; Ren et al., 2011; Zuraimi et al., 2009). According to the Portuguese legislation (Portaria 353-A/2013), *Chrysonilia* sp. exceeded the limit value of 50 CFU.m<sup>-3</sup> in indoor air samples. All of these fungal species were defined as sources of allergens, inducers of IgE-mediated sensitization and causes of atopic respiratory diseases like allergic rhinitis or asthma (Klarić et al., 2012).

With respect to *Aspergillus* genus three different fungal species in indoor air were detected in a total of 256 isolates. *A. candidus* was the species most frequently isolated (63%), followed by *A. fumigatus* (15%) and *A. niger* (13%). With regard to qualitative assessment of fungal contamination, it is postulated that among other species, *Aspergillus fumigatus*, *Aspergillus versicolor* and *Penicillium* species, all of which were isolated in this study, need to be considered as indicators of humidity problems and/or a potential risk to health.

According to the American Industrial Hygiene Association (AIHA, 1996), determination of biological contamination in environmental samples with the species *Stachybotrys chartarum*, *A. versicolor*, *A. flavus*, *A. fumigatus* and *A. niger* requires implementation of corrective measures (NT-SCE-02, 2009; AIHA 1996). In this study, the 4 species of Aspergillus mentioned above were identified in the bedroom and living-room from ECC 1 for *A. versicolor*, living-room from ECC 1 and ECC 2 for *A. fumigatus*, living-room from ECC 2 for *A. niger* and canteens from ECC 3 for *A. flavus*.

Aspergillus fumigatus was found in 2 indoor spaces: living-room from ECC 1 and ECC 2 and in 1 outdoor sample. Hospital guidelines for ECC fungal contamination evaluation were adopted considering the occupants susceptibility, since these guidelines are more demanding than Portuguese law. Faure et al., (2002) used the acceptability threshold for hospital settings > 2 CFU.room<sup>-1</sup> without A. fumigatus. This threshold was used to extrapolate air results and perform corrective measures in the contaminated areas. Considering this threshold, 12.5 % of the indoor air samples presented species from A. fumigatus. Table 2.7 also shows that in indoor surfaces samples 14 different fungal species were isolated in a total of 405,000 isolates. Species from Penicillium genera were the ones most frequently isolated (22%), followed again by Aspergillus sp. (17%), Chrysosporium sp. (12%), Cladosporium sp. (10%) and Chrysonilia sp. (10%). Other fungi were also isolated including: Acremonium sp., Scopulariopsis sp., Scytalidium sp., Geotrichum sp., Alternaria sp., Syncephalastrum sp. and Paecilomyces sp. Among Aspergillus genus, A. candidus was found on surfaces (14%), being the A. niger (86%) the most isolated species.

Table 2.8 - Most Frequent Fungi Genus Isolated Indoor

<b>Indoor Surfaces Samples</b>	Frequency (n; %)
Penicillium sp.	90000; 22
Aspergillus sp.	70000; 17
Chrysosporium sp.	50000; 12
Cladosporium sp.	40000; 10
Chrysonilia sp.	40000; 10
Others	115000; 28
Indoor Air Samples	Frequency (n; %)
Penicillium sp.	600; 38
Aspergillus sp.	256; 16
Chrysonilia sp.	224; 4.2
Others	493; 41
Outdoor Air Samples	Frequency (n; %)
Penicillium sp.	84; 26
Aspergillus sp.	64; 20
Chrysonilia sp.	48; 15
Others	122; 38

The role of human occupancy as a source of fungi is poorly understood. Therefore, an additional approach was applied in ECC 1, in order to better understand the possible influence of human occupancy on fungi contamination. Bedroom and living-room were studied on 3 consecutive days during the occupied and non-occupied period. According to Figure 2.14, the indoor fungal load was always lower in bedroom before occupancy than after occupancy. In living-room the highest values were measured before occupancy, with the exception for the living-room(c) that presented a high fungal load after occupancy. However, in that case outdoor fungi concentration also presented high levels which may explain this phenomenon. Outdoor fungal load was always higher than indoor concentration, with exception for living-room(a) before occupancy (132 CFU.m<sup>-3</sup>).

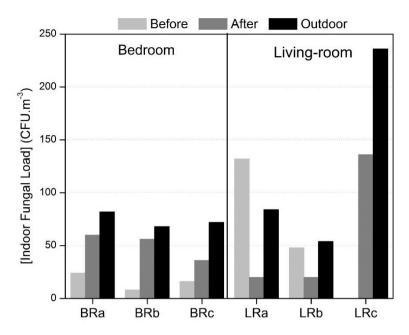


Figure 2.14 – Air fungal load assessed before and after elderly occupancy (values in CFU.m<sup>-3</sup>).

Figure 2.15 shows the most frequent fungi isolated before and after elderly occupancy in bedroom and living-room. In bedroom, *Chrysonilia* sp. (26%) was the most common, followed by *Cladosporium* sp. (21%) before occupancy. *Penicillium* sp. and *Chrysonilia* sp. were the most identified after occupancy, both with 36%. In living-room *Penicillium* sp. (33%) was also the most isolated before occupancy and *Neoscytalidum* sp. the most abundant species after elderly occupancy (44%). Further, other fungi were isolated including *Acremonium* sp., *Stachybotrys chartarum*, *Chrysosporium* sp. and *Geotrichum* sp. among *Aspergillus* genus, *A. fumigatus* and *A. niger* were also identified.

Stachybotrus chartarum was isolated in bedroom, such as A. fumigatus, grow in environments with high levels of humidity and produces mycotoxins, including macrocyclic trichothecenes and satratoxin G (Urvashi et al., 2011). Inhalation of S. chartarum has been associated with multiple symptoms including muscle aches, headaches, cough, pulmonary hemorrhage, dermatitis and interstitial lung disease. It is important to be aware to the fact that the fungal load found for this species (12 CFU.m<sup>-3</sup>) might be underestimated since this fungus is not easily aerosolized because of its sticky nature becoming difficult to be detected by air sampling (Duchaine & Meriaux, 2001). In addition, S. chartarum has a slow growth and thus difficult to detect by conventional methods (Malta-Vacas et al. 2012; Cooley et al. 1998). The presence of these species, and also the identification of species belonging to Aspergillus genus, requires immediate intervention in order to avoid the health risk of elderly to infections (NT-SCE-02, 2009; AIHA, 1996).

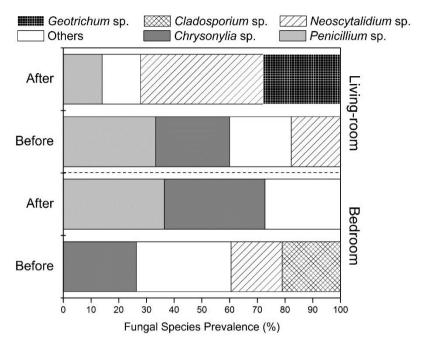


Figure 2.15 – Most frequent fungi isolated before and after elderly occupancy.

Culture methods have several disadvantages including poor precision and a highly variable underestimation of exposure. The underestimation depends on sampling strains, microbial robustness, and size of aggregates that may grow only into one colony. Further, the number of CFU depends on culture conditions, nutrient medium, and presence of other species. Consequently, results based on cultivation are at best semi-quantitative (Eduard & Halstensen, 2009, Eduard & Heederik 1998). However, in the case of a possible fungal exposure through inhalation, conventional methods offer the advantage of enabling identification and quantification only of viable microorganisms and; subsequently, the ones producing higher risk for occupant health (Samson et al., 2000). Therefore, conventional and molecular biological methods when applied together are complementary tools useful in the evaluation of microbiological contamination (Viegas et al., 2012; Malta-Vacas et al., 2012).

## Fungal assessment by molecular methods

Molecular methods were applied in order to detect the presence of *Aspergillus fumigatus* complex. Species from this complex belong to the group of microorganisms considered indicators of moisture-damaged buildings (Samson et al., 1994). Their spores are easily spread in the air and, therefore, pose a high risk of exposure for both animals and humans

(Land et al., 1987). Moreover, the conidia from these species are small enough to traverse the terminal respiratory airways and reach the pulmonary alveoli (Ben-Ami et al., 2010).

Real-time PCR did not detect the presence of *A. fumigatus* complex probably due to the presence of environmental contaminants indoors that inhibit PCR analyses, which may result in false-negative samples (Burton et al., 2008). Another explanation may be the fact that the number of microorganisms present in the environment was below the threshold of amplification by real-time PCR. This is in agreement with the low levels of fungal load detected by conventional methods. One cannot exclude that there is underestimation of fungal load by conventional methods as Aspergillus is a termophilic species (Fulleringer et al., 2006) and all collected samples were incubated at 27°C.

# 2.4.6 Daily average exposure

Daily average exposure for each elder (E<sub>i</sub>) was assessed by integrating the results obtained from the time-budget survey (Sub-chapter 2.4.1) with the pollutants concentrations measured in the different micro-environments. Hence, the Equation 1 was applied:

$$E_{i} = \frac{\sum_{j=1}^{m} c_{ij} \cdot t_{ij}}{\sum_{j=1}^{m} t_{ij}}$$
(2.1)

Where  $C_{ij}$  is the concentration of the pollutant measured in the  $j^{th}$  micro-environment of the  $i^{th}$  individual,  $t_{ij}$  is the time spent by the  $i^{th}$  individual in the  $j^{th}$  micro-environment. The total number of micro-environments is m such that:

$$\sum_{j=1}^{m} t_{ij} = 24 h$$

# 2.4.6.1 Exposure to CO, CO<sub>2</sub>, VOC, PMx in 10 ECCs

Table 2.9 and Figure 2.16 present the absolute and the relative daily average exposure to air pollutants assessed in the ten selected ECCs.

Carbon dioxide daily average exposure varied between 473 mg.m<sup>-3</sup>.h<sup>-1</sup> in ECC 9 (the ECC that presented the highest ventilation rate) and 2,132 mg.m<sup>-3</sup>.h<sup>-1</sup> in ECC 6. In all ECCs, bedrooms had the highest contribution to the CO<sub>2</sub> exposure (70% in average) due to the fact that this pollutant was produced inside the bedrooms during their occupancy, spaces were

smaller, mechanical ventilation was inexistent or not used and doors and windows were closed during the night.

In all ECCs, except for ECC 10, the contribution of bedrooms for CO was greater than 60%. Once this pollutant is associated with combustion processes and was not produced inside the bedrooms, this fact can be explained by the existence of air extractions in the bedroom's toilets that promoted lower pressures in these spaces and the transport of air and pollutants from the ECCs spaces to the toilets passing through the bedroom. The daily CO exposure varied between 0.02 mg.m<sup>-3</sup>.h<sup>-1</sup> in ECC 9 and 2.3 mg.m<sup>-3</sup>.h<sup>-1</sup> in ECC 10.

Toward VOC daily average exposure, the values varied from  $0.06~\text{mg.m}^{-3}.\text{h}^{-1}$  in ECC 7 to  $1.2~\text{mg.m}^{-3}.\text{h}^{-1}$  in ECC 5 and 6, respectively. In the case of this pollutant the relative contribution of the different micro-environments depended on the ECC. While in ECC 7 the bedroom contribution was 100%, because the concentration measured in the other micro-environments was lower than the detection limit of the equipment, in ECC 10 the bedroom contribution was almost null due to the low concentrations measured in this space  $(0.0004~\text{mg.m}^{-3})$ .

Table 2.9 – Daily average exposure assessed for each ECCs for VOC, CO<sub>2</sub>, CO, PM1, PM2.5 and PM10. VOC, CO<sub>2</sub> and CO are presented in mg.m $^{-3}$ .h $^{-1}$  and PM10, PM2.5 and PM1 are presented in  $\mu$ g.m $^{-3}$ .h $^{-1}$ .

		VOC	CO <sub>2</sub>	CO	PM1	PM2.5	PM10
	BR	0.59	852	0.16	0.80	1.9	5.6
_	LR	0.12	238	0.04	0.29	0.91	3.4
ECC 1	Out	0	24	0	0	0	0.79
$\Xi$	Others	0.11	212	0.03	0.26	0.81	3
	Total	0.82	1326	0.23	1.4	3.6	13
-	BR	0.06	889	0.04	1.4 1.2	2.5	7.3
7	LR	0.05	252	0.03	0.80	1.5	4.3
ECC 2	Out	0.00	10	0	0	0	0.27
$\Xi$	Others	0.03	177	0.02	0.56	1.1	3.0
	Total	0.14	1328	0.09	2.5	5.1	15
	BR	0.44	1089	0.54	2.5 1.6	3.0	11
$\alpha$	LR	0.04	298	0.04	0.95	1.5	4.5
ECC 3	Out	0	0	0	0	0	0
函	Others	0.02	121	0.02	0.38	0.59	1.8
	Total	0.50	1508	0.60	2.9	5.1	17
	BR	0.16	1400	0.26	3.9	6.9	14
4	LR	0.08	345	0.07	0.89	2.1	5.9
ECC 4	Out	0	6	0	0	0	0.15
函	Others	0.05	231	0.05	0.60	1.4	4.0
	Total	0.29	1982	0.38	5.2	10	24
	BR	0.40	1200	0.65	1.00	1.57	3.0
ECC 5	LR	0.05	898	0.51	2.92	2.64	11
$\mathcal{C}$	Out	0	0	0	0	0	0
页	Others	0	0	0	0	1.26	0
	Total	0.45	2098	1.2	3.9	5.5	14
	BR	0.44	1350	0.73	0.97	1.6	3.4
9	LR	0.03	493	0.28	1.7	2.4	6.2
ECC 6	Out	0	0	0	0	0	0
Ш	Others	0.02	289	0.16	1.0	1.4	3.7
	Total	0.49	<b>2132</b> 1073	1.2	3.7	5.4	13
	BR	0.05		0.06	6.42	2.35	19
ECC 7	LR	0	271	0	2.38	3.18	17
$\mathcal{Z}$	Out	0	9	0	0.16	0.08	2.6
Щ	Others	0	17	0	0.15	1.15	1.0
	Total	0.05	1370	0.06	9.1	6.8	40
	BR	0.46	487	0.33	2.2	4.9	8.8
ECC 8	LR	0.05	246	0.04	1.2 0.20	2.7	5.8
ğ	Out	0.02	13	0.01	0.20	1.4	4.1
щ	Others	0.01	60	0.01	0.12	0.66	1.4
-	Total	0.54	806	0.39	3.7	9.7	20
	BR	0.02	368	0.20	1.6	2.7	5.9
ECC 9	LR	0	105	0.14	1.4	1.8	5.0
Ö	Out	0	0	0	0	0	0
П	Others	0	0	0	0	0	0
	Total	0.02	473	0.34	3.0	<b>4.5</b> 19	11
0	BR	1.1	1089	0	16		26
ECC 10	LR	0.93	553	0.23	1.3	3.9	6.1
$\Sigma$	Out	0.01	3	0 0.08	0.06	0.14	0.28
Щ	Others	0.33	194		0.45	1.4	2.1
TOT	Total	2.4	1839	0.31	18 5.3	24	35 20
TOT	AL	0.57	1486	0.48	5.3	8.0	20

For the particles, the living-room contribution was higher compared to the previous pollutants (on average 37%, 33% and 34% for PM10, PM2.5 and PM1, respectively). This is explained by the fact that in living-room there were more people, including elders and supporters, which promote the re-suspension of particles. The highest PM10 exposure was measured in ECC 7 (39 µg.m<sup>-3</sup>.h<sup>-1</sup>). This ECC presented the highest outdoor PM10 concentrations that not only had an impact on the outdoor PM10 exposure but also on the indoor exposure due to the infiltration of particles to the indoor.

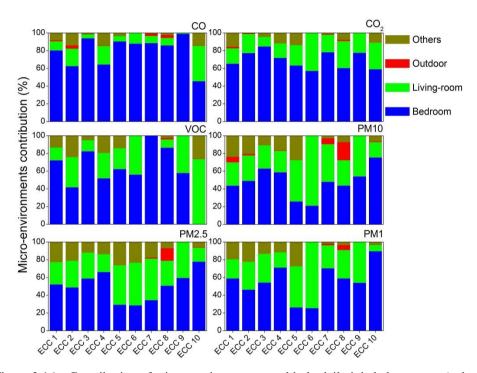


Figure 2.16 – Contribution of micro-environments to elderly daily inhaled exposure (values in %).

## 2.4.6.2 Exposure to PM10 and its components in 4 ECCs

Table 2.10 and Figure 2.17 present the absolute and the relative daily average exposure to air pollutants assessed in the four selected ECCs, considering PM10 and its components.

PM10 daily average exposure varied between 11  $\mu$ g.m<sup>-3</sup>.h<sup>-1</sup> in ECC 3 and 16  $\mu$ g.m<sup>-3</sup>.h<sup>-1</sup> in ECC 4, which is located in an urban area. For the components that presented higher contributions of indoor sources – OC and  ${\rm CO_3}^{2^-}$  – similar daily exposure levels were registered in all ECCs, irrespective of their localization (3.4 and 1.4  $\mu$ g.m<sup>-3</sup>.h<sup>-1</sup>, respectively). The contribution of the bedrooms for those compounds was predominant (57% for both of them).

Higher daily exposure levels were registered in ECC 4 for the elements As, Fe, K, Sb, Sc and Sm (0.59, 150, 190, 0.97, 0.015 and 0.012 ng.m<sup>-3</sup>.h<sup>-1</sup>, respectively). These elements presented I/O ratios lower than one indicating their association with outdoor sources. This ECC was more affected by traffic, as it was located in an urban area, near to a highway and to International Airport of Lisbon (see details in chapter 3). For the above mentioned elements, the contribution of the different micro-environments to the daily average exposure was highly dependent on the ECC.

Table 2.10 – Daily average exposure assessed for each ECCs for PM10, carbonaceous compounds and trace elements. PM10 and carbonaceous compounds are presented in  $\mu g.m^{-3}.h^{-1}$  and trace elements are presented in  $ng.m^{-3}.h^{-1}$ .

		PM10	OC	EC	CO <sub>3</sub> <sup>2-</sup>	As	Co	Cr	Fe	K	Na	Sb	Sc	Sm	Zn
	BR	7.8	2.6	0.44	0.91	0.07	0.02	3.6	20	52	140	0.23	0.002	0.002	6.6
_	LR	2.8	0.60	0.17	0.26	0.05	0.01	3	14	16	120	0.07	0.001	0.001	7.7
ECC	Out	0.71	0.06	0.02	0.02	0.01	0.00	0.71	6	6	42	0.03	0.001	0.001	0.97
函	Other	2.5	0.56	0.16	0.23	0.38	0.01	2.7	12	14	110	0.06	0.001	0.001	6.9
	Total	14	3.8	0.79	1.4	0.51	0.045	10	52	88	412	0.39	0.005	0.005	22
	BR	4.9	1.7	0.42	0.98	0.06	0	2.70	16	44	120	0.23	0.001	0.000	7.1
2	LR	3.8	0.83	0.33	0.33	0.06	0.03	3.20	55	44	69	0.31	0.005	0.003	10
ECC	Out	0.26	0.03	0.01	0.02	0.002	0.001	0.11	3	1.9	12	0	0	0	0.32
函	Other	2.6	0.58	0.23	0.23	0.04	0.02	0.22	40	31	49	0.22	0.003	0.002	7.3
	Total	12	3.1	1.0	1.6	0.16	0.05	6.2	114	121	250	0.76	0.009	0.005	25
	BR	6.8	3	0.49	0.87	0.09	0.06	0.5	30	47	81.00	0.25	0.004	0.002	26
3	LR	3.3	0.79	0.34	0.29	0.05	0.02	1.9	60	27	57.00	0.47	0.002	0.001	7.5
ECC	Out	0	0	0	0	0	0	0	0	0	0	0	0	0	0
Щ	Other	1	0.32	0.14	0.12	0.02	0.001	0.8	25	11	23.00	0.19	0.001	0.001	3.1
	Total	11	3.7	1.0	1.3	0.17	0.08	3.2	115	85	161	0.9	0.007	0.004	37
	BR	7.9	1.1	0.44	0.49	0.39	0.04	10	50	97	110	0.51	0.003	0.002	25
4	LR	4.8	1.1	0.29	0.58	0.12	0.03	2.6	56	40	100	0.26	0.007	0.006	7.7
ECC	Out	0.22	0.02	0.01	0.02	0.01	0.002	0.10	3.7	20	23	0	0	0	4
Й	Other	3.2	0.77	0.19	0.04	0.08	0.02	1.7	38	30	67	0.18	0.005	0.004	5.2
	Total	16	3.0	0.93	1.1	0.60	0.09	14	148	187	300	0.95	0.015	0.012	42
	Ei	13	3.4	0.92	1.3	0.36	0.07	8.5	107	120	281	0.75	0.009	0.007	31

Chromium and Zn (the elements with the highest ratio I/O) presented daily average exposure of 10 and 30 ng.m<sup>-3</sup>.h<sup>-1</sup>, respectively. These elements had a similar behaviour: higher contribution of bedrooms from ECC 3 and ECC 4 and greater exposure levels from the living-room of ECC 2.

In the bedroom of ECC 2 the concentration of Sm and Co was always below the detection limit  $(1.6 \text{ x } 10^{-3} \text{ and } 7.6 \text{ x } 10^{-2} \text{ ng.m}^{-3}.\text{h}^{-1}$ , respectively), and for this reason the daily exposure was null.

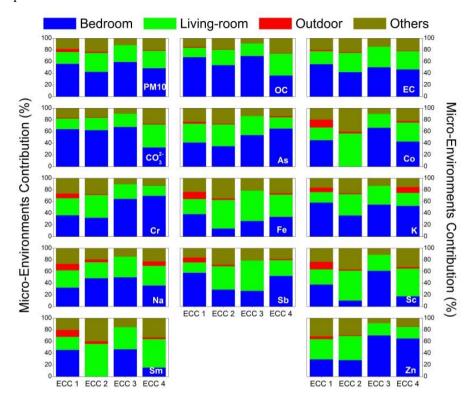


Figure 2.17 – Contribution of micro-environments to elderly daily exposure by PM10 and its components (values in %).

## 2.4.7 Daily average inhaled dose

The daily average inhaled dose for each elder  $(E_{di})$  was assessed by integrating the time spent in each micro-environment, the concentration of the pollutants for the period of interest, the inhalation rate (IR) and the body weight (BW) according Equation 1.3:

$$E_{di} = \frac{\sum_{j=1}^{m} (c_{ij} \cdot t_{ij} \cdot IR_{ij})}{\sum_{j=1}^{m} t_{ij}}$$
(1.3)

The IR's associated with the three different micro-environments – bedroom, living-room and outdoor – were recommended by U.S. EPA (2011) for people with more than 61 years old in three distinct activities – sleep, sedentary and light intensity, respectively. These values were selected to be used as the recommended inhalation rates since they were based on three studies: U.S. EPA (2009), Stifelman (2007) and Brochu et al. (2006). Table 2.11 presents the inhalation values used in this study. The body weight used in this work was 80 kg, also based on U.S. EPA (2011). Although several studies showed that inhalation rate depends on body weights, energy expenditure rate, oxygenation rates, pollutants concentration in each micro-environment, time activity pattern, etc. (Lazaridis & Colbeck, 2010; Brochu et al., 2014), in this work it was not possible to achieve the specific body weight of each elder' voluntary. The daily average inhaled dose was calculated for PM10, carbonaceous components and trace elements for each individual.

Table 2.11 – Inhalation rates used in this study. Based on U.S. EPA (2011).

Activity	Age Group	Inhalation Rate (m³.min⁻¹)
	61 to < 71	0.0052
Sleep	71 to < 81	0.0053
Sieep	≥ 81	0.0052
	Mean	0.0052
	61 to < 71	0.0049
Sedentary	71 to < 81	0.005
Passive	≥ 81	0.0049
<del>-</del>	Mean	0.0049
	61 to < 71	0.0012
Light	71 to < 81	0.0012
Intensity	≥ 81	0.0012
	Mean	0.0012

## 2.4.7.1 Inhaled dose of CO, CO<sub>2</sub>, VOC, PMx

Table 2.12 and Figure 2.18 present the absolute and the relative daily average dose inhaled to air pollutants assessed in the ten selected ECCs.

Carbon monoxide daily average inhaled dose varied from 0.002 mg.kg<sup>-1</sup> to 0.031 mg.kg<sup>-1</sup> in ECC 9 and ECC 1, respectively. As it was observed in the exposure levels, higher inhaled dose were registered in bedrooms. CO<sub>2</sub> daily average inhaled dose varied between 44

 $mg.kg^{-1}$  in ECC 9 and 197  $mg.kg^{-1}$  in ECC 5. In all ECCs, bedrooms had the highest contribution to the  $CO_2$  inhaled dose (68% in average).

Considering the VOC, the ECC 7 presented the lowest daily average inhaled dose (0.006  $\mu g.kg^{-1}$ ), with 100% contribution of the bedroom. ECC 6 presented the highest daily average inhaled dose of 0.106 mg.kg<sup>-1</sup>. In average, bedroom and living-room had a contribution of 63% and 27%, respectively.

Table 2.12 – Daily average inhaled dose assessed for each ECCs for VOC, CO<sub>2</sub>, CO, PM1, PM2.5 and PM10. VOC, CO<sub>2</sub> and CO are presented in  $mg.kg^{-1}$  and PM1, PM2.5 and PM10 are presented in  $\mu g.kg^{-1}$ .

		VOC	CO <sub>2</sub>	CO	PM1	PM2.5	PM10
	BR	0.054	78	0.027	0.074	0.17	0.52
-	LR	0.010	21	0.002	0.025	0.078	0.29
ECC 1	Out	0.000	5	0.000	0.000	0.000	0.17
$\Xi$	Others	0.009	18	0.002	0.023	0.070	0.26
	Total	0.073	122	0.031	0.12	0.32	1.2
	BR	0.006	84	0.004	0.111	0.24	0.68
2	LR	0.004	22	0.002	0.071	0.14	0.38
ECC 2	Out	0.000	2.2	0.001	0.000	0.000	0.06
Щ	Others	0.003	16	0.002	0.050	0.096	0.27
	Total	0.013	124	0.008	0.23	0.47	1.4
	BR	0.040	100	0.051	0.146	0.27	0.99
$\epsilon$	LR	0.003	26	0.004	0.082	0.13	0.39
ECC 3	Out	0.000	0	0.000	0.000	0.000	0.00
Щ	Others	0.001	10	0.002	0.033	0.051	0.16
	Total	0.045	136	0.056	0.26	0.45	1.5
	BR	0.015	78	0.024	0.346	0.65	1.3
4	LR	0.007	21	0.006	0.079	0.19	0.53
ECC 4	Out	0.000	5.0	0.000	0.000	0	0.03
Щ	Others	0.005	18	0.004	0.053	0.12	0.35
	Total	0.026	122	0.035	0.48	0.96	2.3
	BR	0.069	127	0.041	0.091	0.15	0.32
ECC 5	LR	0.025	44	0.003	0.153	0.22	0.56
ğ	Out	0.000	0	0.000	0.000	0	0.00
Щ	Others	0.014	26	0.001	0.089	0.13	0.32
	Total	0.11	197	0.046	0.33	0.50	1.2
	BR	0.061	113	0.038	0.094	0.14	0.28
ECC 6	LR	0.045	80	0.005	0.259	0.41	1.0
Ö	Out	0.000	0	0.000	0.000	0	0.00
ш	Others	0.000	0	0.000	0.000	0	0.00
	Total	0.106	193	0.042	0.35	0.55	1.3
	BR	0.006	101	0.005	0.604	0.91	1.8
ECC 7	LR	0.000	24	0.000	0.211	0.68	1.5
$\Xi$	Out	0.000	1.9 1.5	0.000	0.035	0.18 0.041	0.57 0.09
	Others <b>Total</b>	0.000	1.3 128	0.000 <b>0.006</b>	0.013		3.9
	BR	0.006 0.030	45	0.042	<b>0.86</b> 0.152	1.8 0.46	0.81
∞	LR	0.003	21	0.004	0.053	0.24	0.50
ECC 8	Out	0.003	2.7	0.004	0.056	0.29	0.86
$\Xi$	Others	0.001	5.2	0.004	0.013	0.058	0.12
	Total	0.036	74	0.052	0.27	1.0	2.3
	BR	0.018	35	0.002	0.153	0.26	0.56
6	LR	0.013	9.4	0.000	0.122	0.16	0.45
ECC 9	Out	0.000	0	0.000	0.000	0	0.00
ы	Others	0.000	0	0.000	0.000	0	0.00
	Total	0.031	44	0.002	0.28	0.42	1.0
	BR	0.000	103	0.100	1.473	1.8	2.5
10	LR	0.021	49	0.083	0.114	0.34	0.54
ECC 10	Out	0.000	0.6	0.001	0.014	0.030	0.06
$\Xi$	Others	0.007	17	0.029	0.04	0.12	0.19
	Total	0.028	169	0.21	1.6	2.3	3.3
TOTA	AL	0.47	131	0.049	0.48	0.88	1.9

For the particles, the daily average inhaled dose was higher in living-rooms, comparing with the other pollutants. For PM1, PM2.5 and PM10 the contribution of the living-room was, on average, 31%, 33% and 35%, respectively.

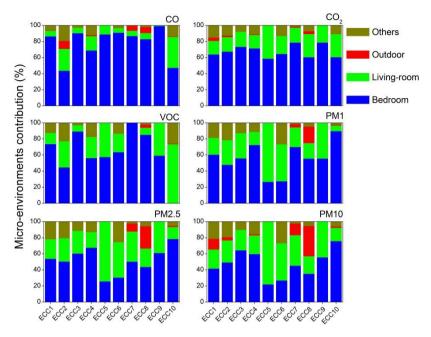


Figure 2.18 – Contribution of micro-environments to elderly daily inhaled dose by air pollutants (values in %).

## 2.4.7.2 Inhaled dose of PM10 and its components

The daily average inhaled dose was calculated for PM10, carbonaceous components and trace elements for each individual. Table 2.13 and Figure 2.19 present the absolute and the relative daily average inhaled dose for the studied air pollutants and for each ECCs.

PM10 daily average inhaled dose varied between 1.02  $\mu g.kg^{-1}$  and 1.46  $\mu g.kg^{-1}$ . OC, EC and CO<sub>3</sub><sup>2-</sup> presented daily average inhaled doses of 0.3, 0.08 and 0.13  $\mu g.kg^{-1}$ , respectively.

Higher inhaled doses were registered in ECC 4 for the elements As, Cr, Fe, K, Sb, Sc, Sm and Zn. With the exception of Cr and Zn, the other elements presented I/O ratios lower than one indicating their association with outdoor sources. As it was described before Cr and Zn presented I/O ratio higher than 1, suggesting an association with an indoor source (Figure 2.12). The ECC4 was more affected by traffic, as it was located in an urban area, near to a highway and to International Airport of Lisbon (see details in chapter 3).

The contribution of each micro-environment for the inhaled dose presented a similar behaviour as the contribution for the exposure and was highly depended on pollutant and on the ECC.

Table 2.13 – Daily average inhaled dose assessed for each ECCs for PM10, carbonaceous compounds and trace elements. PM10 and carbonaceous compounds are presented in  $\mu g.kg^{-1}(x10E^{-4})$  and the trace elements are presented in  $ng.kg^{-1}(x10E^{-4})$ .

		PM10	OC	EC	$CO_3^{2-}$	As	Co	Cr	Fe	K	Na	Sb	Sc	Sm	Zn
	BR	740	250	41	86	4.8	2.1	350	1900	5000	12750	22	0.19	0.21	620
1	LR	250	60	15	23	0.7	0.96	270	1200	1430	11000	6.3	0.12	0.1	690
ECC	Out	2.6	0.21	0.06	0.06	0.03	0.023	1.6	22	22	152	0.11	0.002	0.002	3.5
Щ	Others	220	50	14	20	0.58	0.86	240	1070	1280	9800	5.6	0.11	0.09	610
	Total	1213	360	70	129	6.1	3.9	862	4192	7732	33702	34	0.42	0.40	1924
	BR	460	160	40	90	5.2	0	250	1500	4100	11600	21	0.1	0	670
7	LR	330	74	30	30	5.2	2.6	280	4900	3900	6100	28	0.42	0.23	920
ECC	Out	0.92	0.09	0.05	0.07	0.007	0.005	0.39	10	6.7	42	0.07	0.0007	0.0007	1.1
Щ	Others	230	50	21	21	3.6	1.9	200	4200	2700	4300	19	0.29	0.16	650
	Total	1021	284	91	141	14	4.5	730	10610	10707	22042	68	0.81	0.39	2241
	BR	640	240	46	83	8.4	5.6	480	2900	4430	7700	23	0.38	0.16	2400
$\tilde{\omega}$	LR	290	70	30	26	4.8	1.9	176	5300	2430	5100	42	0.16	0.12	670
ECC	Out	0	0	0	0	0	0	0	0	0	0	0	0	0	0
Щ	Others	120	30	12	11	1.9	0.77	71	2200	990	2060	17	0.07	0.05	270
	Total	1050	340	88	120	15	8.3	727	10400	7850	14860	82	0.61	0.33	3340
	BR	740	100	41	46	36	3.6	950	4700	9100	10200	48	0.25	0.18	2350
4	LR	430	100	26	51	10	2.5	230	5000	3600	9000	23	0.64	0.52	680
ECC	Out	0.79	0.08	0.03	0.07	0.05	0.009	0.36	13	70	81	0.08	0.0008	0.001	1.3
Щ	Others	290	70	18	34	6.9	1.7	150	3330	2400	6000	16	0.43	0.35	460
	Total	1461	270	85	131	53	7.8	1330	13043	15170	25281	87	1.3	1.1	3491
	Ed <sub>i</sub>	1186	314	84	130	22	6.1	912	9561	10365	23971	68	0.79	0.54	2749

As far as we know, this work generated the first data on particles components inhaled dose for indoor residential and service buildings. Therefore, it is not possible to compare the inhaled doses measured in this study with results obtained for other indoor environments and populations.

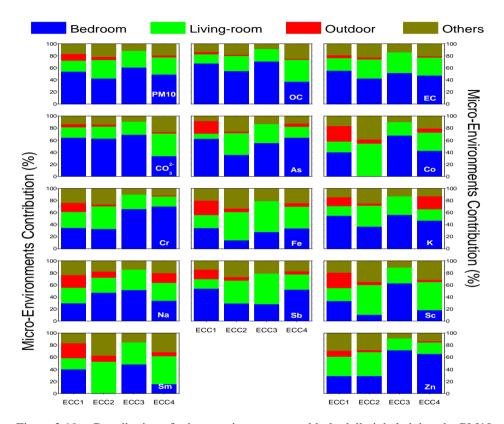


Figure 2.19 – Contribution of micro-environments to elderly daily inhaled dose by PM10 and its components (values in %).

# 2.4.8 Deposited surface area of nanoparticles in elderly lungs

The deposited surface area of nanoparticles in elderly lungs was determined in both indoor micro-environments where elders spent the most of their time: bedroom and living-room.

Table 2.14 reports the deposited surface area measured by the NSAM, which ranged from  $10 \ \mu m^2 .cm^{-3}$  to  $46 \ \mu m^2 .cm^{-3}$ . The application of the Mann Whitney test showed that living-rooms presented significantly higher results comparing with the assessed bedrooms (p = 0.00) and it was the bedroom of ECC 2 that presented the lowest levels of exposure considering the other assessed bedrooms. In fact, it is important to consider that the

cleaning of these micro-environments was made by dry processes instead of wet processes, which could promote re-suspension of particles. In ECC 2 the periodicity of cleanness was only once a week, which can, probably, explain the low values of deposited nanoparticles. Living-room from the ECC 3 presented the highest value of deposited surface area (46  $\mu$ m<sup>2</sup>.cm<sup>-3</sup>).

T.1.1. O 14	<ul> <li>Deposited Surface</li> </ul>	A (DCA)		1 1.	
19NIP / 14 -	- Denostrea Surrace	Area (IIXA)	i in elderiv	llings ny	i nanonariicies
1 4010 2.17	Deposited Duriace	I II Ca (DDII)	, iii ciacii y	Tungo U	manoparticios.

Micro-	environments	Exposure time <sup>(a)</sup>	Average DSA (µm².cm⁻³) <sup>(b)</sup>
ECC 1 -	Bedroom	36h	26
ECC I -	Living-Room	36h	19
<b>500.0</b>	Bedroom	24h	10
ECC 2 -	Living-Room	36h	24
ECC 3	Bedroom	36h	14
ECC 3	Living-Room	36h	46
	Bedroom	24h	23
ECC 4	Living-Room	36h	38

a) Exposure time occurs during 3 consecutive occupied periods; b) Considering an average lung area of  $80 \text{ m}^2$ .

Table 2.15 aimed to compare the deposited surface area measured in this study with results determined by different authors in different environments. Results showed that deposited surface area obtained in the current study were lower comparing with the majority of the other studies. Considering the occupational studies only the Avian Base study presented lowest deposited surface area (Buonanno et al., 2012), probably due to the fact that this work was performed in outdoor environments. The results related to cooking activities presented quite similar results between themselves, even considering different kind of cooking activities (Bordado et al., 2012; Buonanno et al., 2010). These results were higher comparing with the current study, because besides the existence of cooking activities in ECCs, they were performed in specific and delimitated areas where elders were not allowed to enter. Therefore, the distance between cooking activities and meeting points was higher in ECCs.

Considering the other susceptible population (children) it is possible to observe that their deposition surface area was much higher comparing the results obtained on this study (Buonanno et al., 2013). Besides both elders and children are considering a susceptible

population the latest are more vulnerable to environmental pollutants since they breathe more air relative to their body weight and also have a lower capacity to deal with toxic chemicals (Firestone et al., 2008; Selgrade et al., 2008).

Table 2.15 – Average Deposited Surface Area (DSA) in different studies.

Authors	Type of Study	Site	DSA (μm².cm <sup>-3</sup> )
This Study	Indoor	Bedroom (ECC)	1.8E1
This Study	Indoor	Living-room (ECC)	3.2E1
Buonanno et al., 2012	Occupational	Avian Base	3.6E0
Wilson et al, 2007	Outdoor	Minneapolis	1-5E1
Albuquerque et al., 2012	Outdoor	Trailer near traffic road	6.6E1
Gomes et al., 2012	Occupational	Tungsten Inert Gas	2.8E2
Bordado et al., 2012	Indoor	Cooking	3.1E2
Buonanno et al., 2010	Indoor	Pizzeria	3.6E2
Gomes et al., 2012	Occupational	Metal Active Gas	7.7E2
Buonanno et al., 2013	Indoor+Outdoor	Children daily exposure	1.3E3
Gomes et al., 2012	Occupational	Friction Stir Welding	1.1E4

In order to characterize and to understand the nanoparticles deposited surface area on alveolar regions of the respiratory system of the elders, its temporal distribution was achieved during the occupied period. Figure 2.20 shows the temporal variation of deposited surface area in elderly lungs evaluated in bedrooms and living-rooms.

The temporal distribution of deposited surface area in elderly lungs assessed in bedrooms showed a pattern throughout the night campaign characterized by a decrease of the DSA. In ECC 1, ECC 2 and ECC 4 peaks associated with the movement of elders and supports.

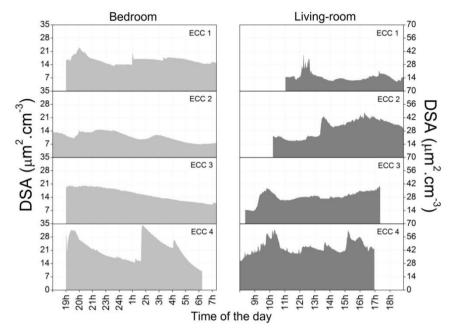


Figure 2.20 – Temporal distribution of Deposited Surface Area in bedrooms and living-rooms of all studied Elderly Care Centers. Results are presented in  $\mu m^2$ .cm<sup>-3</sup>.

In living-rooms, it was possible to observe higher DSA values comparing with the bedrooms and this phenomenon was associated with the entrance and exit of the old people in living-room, increasing the movement and the possibility of particles re-suspension. In detailed, the ECC 1 presented one high peak at 12 h which correspond to the lunch time. This peak could be explained by two factors: 1) the increase of movement by elders and supports due to the moving to the canteen and 2) the transport of particles from the cooking activities into living-room. Both reasons may justify the increased of deposited surface area in elderly lungs. In the living-room of the ECC 3 most elders were dependent and, therefore, they got into living-room in the morning, between 9 h to 10 h, and stayed there all day, even during the lunch time.

## 2.5 Limitations

The present work studied the institutionalized elderly exposure and dose to particle components. A couple of limitations were identified during the execution of this work:

a) The first limitation was related to the lack of information about the topic. Despite the importance of healthy air in Elderly Care Centers, IAQ studies have been focused mainly on schools (e.g. Canha et al., 2014a; Canha et al, 2013; Canha et al., 2012a; Pegas et al., 2011a,b; Canha et al., 2011; Pegas et al., 2010; Canha et al, 2010); homes (e.g.

Osman et al., 2007), offices (e.g. Bluyssen et al., 1996) and other indoor microenvironments (e.g. Ramos et al., 2014; Viegas et al., 2014). Some of these studies have shown evidence that indoor air pollution increases the risk of respiratory and atopic diseases, but information on health effects of such pollutants in the elderly is scarce;

- b) The second limitation was associated with the difficulty to get the support from elderly people. Some elders no longer have their best faculties, being very challenging to: 1) speak with them; 2) understand their concerns; 3) explain to them why a group of unfamiliar people were inside their bedrooms; or 4) clarify that the sampling equipments were not dangerous. This is the reason why only were studied 4 ECCs instead of the 10 ECCs that were selected in a previously study (Almeida-Silva et al., 2014a);
- c) Finally, the last limitation was associated with the impossibility to perform measurements near their breathing zone, which is the best approach to calculate the human exposure to air pollutants. Nevertheless, non-significant biases were involved in this procedure, since the majority of institutionalized elders do not approached to indoor emissions sources. Truly, they do not cook, do not clean and rarely do something different than stay in the bedroom or living-room.

## 2.6 Conclusions

Results from this work clearly showed that exposure to indoor air pollutants is a current problem that is necessary to be aware.

In general, the developed time-budget survey showed that elders living in ECCs spent 95% of their time indoors and this value increased in institutions with higher number of bedridden.

Considering the results of air pollutants it was possible to conclude that:

- In almost all ECCs, living-rooms had lower Q<sub>1</sub> values due to higher number of people that occupied these spaces and 30% and 100% of the total indoor micro-environments evaluated did not meet the Portuguese legislation for bedrooms and living-rooms, respectively;
- 40% of the studied bedrooms exceeded the CO<sub>2</sub> limit values provided from Portuguese legislation;
- CO<sub>2</sub> and CO average concentrations in bedrooms were higher than in livingrooms;
- O<sub>3</sub> presented low concentrations in all ECCs;
- CO and CH<sub>2</sub>O did not exceeded the Portuguese limit values;
- The Portuguese limit value for VOC was exceeded in 75% of the analysed indoor micro-environments:
- In average, PMx concentrations in living-rooms were significantly higher than in bedrooms, due to the PM re-suspension;

- PM10 indoor concentrations neither exceed the Portuguese guideline for indoor air quality nor the reference value of 50 μg.m<sup>-3</sup> established by the World Health Organization;
- OC, EC and CO<sub>3</sub><sup>2-</sup> represented 47% of the total PM10 mass measured by gravimetry;
- OC, EC and CO<sub>3</sub><sup>2</sup>- concentrations were significantly higher indoors;
- Only Cr and Zn presented I/O ratio higher than 1;
- Penicillium and Aspergillus genera were the most abundant in air and surfaces;
- The species A. fumigatus was present in 12.5% of the total indoor micro-environments assessed;
- Fungal load in bedroom was higher after elderly occupancy, but living-room presented lower amounts of fungi after occupancy;
- 37.5% of the assessed micro-environments presented higher indoor fungal concentrations than outdoor;
- Bedrooms had the highest contribution to the CO<sub>2</sub> exposure and inhaled dose (70% and 68%, respectively);
- Daily CO exposure varied between 20E-02 mg.m<sup>-3</sup> and 23E-01 mg.m<sup>-3</sup>;
- The relative contribution of different micro-environments to VOC depends on the ECCs;
- Living-room contribution to exposure and to inhaled dose was higher for particles, compared to the other pollutants;
- Bedrooms were the micro-environment that most contributed to the PM10 exposure and inhaled dose (both 52%);
- PM10 daily average exposure varied between 11E00 μg.m<sup>-3</sup> and 16E00 μg.m<sup>-3</sup> and the daily inhaled dose varied between 1.76E-02 μg.kg<sup>-1</sup> and 1.74E-01 μg.kg<sup>-1</sup>;
- The contribution of each indoor micro-environment for the exposure and inhaled dose depended a lot on the particle constituents and on their respective sources;
- The nanoparticles deposited surface area ranged from  $10 \mu m^2 \cdot cm^{-3}$  to  $46 \mu m^2 \cdot cm^{-3}$  and were significantly higher in living-rooms than in bedrooms.

Results also showed that besides living in the same area, the exposure and the inhaled dose of the studied elders differed significantly. The results of this work showed the importance of individual exposure assessment, in order to provide information for the protection of public health, especially for elders who represent one of the most vulnerable groups in society. This approach allowed the identification of the micro-environments with highest impacts on human exposure and proved to be an essential tool to identify health risks, set and review air quality standards and evaluate effective policy interventions.

Additionally, this work showed that the assessment of the integrated exposure to PM components was determinant to accomplish the dose inhaled by elders living in ECCs. In order to compare the personal particle dose with a threshold, an accurate dose evaluation

(approaching as much as possible the actual exposure) should be carried out. This is a crucial aspect, which can only be solved through the assessment of a daily integrated exposure that is able to measure particle concentrations received by people in every microenvironment they visit during a typical day, and by estimating the corresponding doses.

Concluding, it is not possible to affirm that all of these air pollutants achieved the lungs of the elders or which kind of diseases will be induced by these amounts of pollutants. However, the mere fact of the existence of these pollutants means that there is a risk of respiratory and/or atopic diseases. Plus, the inexistence of standards specific for this susceptible population may lead to a misinterpretation of legal compliance, jeopardizing the health and well-being of this population.

# CHAPTER III. ELDERLY EXPOSURE AND DOSE TO PARTICLES – MODELLING APPROACH

## 3.1 Abstract

Elderly population is considered susceptible and, consequently, more vulnerable to possible exposure to air pollutants. The aim of this chapter was to link the external exposure, assessed for elders living in ECCs and autonomous elders, to the internal dose in respiratory tract using a computational model that estimates transport and deposition of particles into the HRT. To achieve the presented goal this chapter was divided in two steps. In first step 384 elders living in 10 Elderly Care Centers were studied. In second step 5 autonomous elders were randomly selected to be part of this study. Different sizes of particulate matter were monitored and a time-activity pattern was applied. In postprocessing, a 1D, mechanistic, respiratory deposition model was used to estimate the elders' particles dose. According to the time-activity survey, old people spent most of their time indoors, such as bedrooms, living-rooms, etc. In general, deposition fraction was higher in male than female for the same activity level and increased with activity level. The results were important to understand the critical areas inside the residences, where elders are most exposed, and to suggest effective mitigation strategies and, so, to reduce their exposure to particles and in extension adverse health effects to this very sensitive age group.

## 3.2 Introduction

Personal integrated exposure to air pollutants is of considerable importance as a key determinant of the pollutants exposure and dose received by an individual and, thus, directly related to health impacts. According to Morawska (2013), up to 30% of the burden of disease from particulate matter exposure can be attributed to indoor-generated particles, indicating that indoor environments are likely to be a dominant factor affecting human health. The assessment of personal exposure to air pollutants is a critical component of epidemiological studies associating air pollution and adverse health effects. Personal exposure to airborne particulate matter can be determined directly by personal measurement and indirectly by ambient measurement at a centrally located site or by micro-environmental models which estimate personal exposure by integrating PM concentrations in micro-environments over time periods people spend there (Almeida-Silva et al., 2015). Individual exposure is quantified for single individuals as they represent some population subgroups, like elders, using estimations based on exposure concentration data and the time of contact.

PM enter into the human body mainly through the inhalation route. Health risk assessment for inhaled particles requires information on local deposition patterns within the human respiratory tract (HRT) and such information can be provided by computational modelling (Morawska et al., 2013). A significant amount of particles may enter into the blood stream and be transferred to the heart and the tissues, potentiate the adverse human health effects (Simkhovich et al., 2008).

Modelling of the particle dynamics and transport can be particularly beneficial in two specific respects: 1) it can provide useful physical insight and enable the interpretation of systems without the need of experiments; and 2) it can be used for parametric investigation and optimization of already developed systems.

The first mathematic model of particle deposition was done in 1935 by Findeisen. Even with several limitations, this model was pioneer establishing the basic norms for the development of other later models (Findeisen, 1935). Since then, several mathematic models has been developed and applied in different fields and proposes, such as, occupational, pharmaceutical, epidemiological and toxicological studies (Pilou et al., 2013; Tena and Clarà, 2012; Mitsakou et al., 2007a,b, 2005; Martonen, 1993; Yeh and Schum, 1980). The use of mathematical models is an advantage on regional dose estimation since, in practice, the regional dose in the respiratory system is very difficult to be addressed experimentally (Hussein et al., 2013).

The human respiratory tract is especially designed, both anatomically and functionally, so that air can reach the most distal areas of the lungs in the cleanest possible conditions. For that, respiratory tract has natural barriers, such as nasal hair, nasal turbinate, vocal cords, the cilia of the bronchial epithelium, the sneeze, etc. Even so, over the years the unintentional and intentional introduction of drugs into the HRT has weakened the protective barriers, enhancing the pollutants to reach the alveoli.

The aim of this work was to link the external exposure, assessed for elders living in ECCs and for autonomous elders, to the internal dose in respiratory tract using a computational model that estimates transport and deposition of particles into the HRT.

# 3.3 Experimental data

In the present work two different approaches were applied: 1) 384 elders living in 10 Elderly Care Centers were studied; and 2) five autonomous elders were randomly selected to be part of this study.

# 3.3.1 First approach – aerosol measurements

In the first approach the PM measures were done in 10 Elderly Care Centers located in Lisbon, Portugal, in collaboration with 384 old people. The voluntaries were institutionalized in those ECCs, which had a range of 7 to 95 occupants per institution and an average of 85 years old. A time-budget survey (TBS) was applied on 384 elders. For this

a close-ended questionnaire was designed, which included information about 1) different activities developed during the day; 2) mealtimes; 3) sleep times and 4) microenvironments where they spend their time. The questionnaire differentiated between time allocation on weekdays and weekends. The questionnaires were applied with the help of collaborators (e.g. socio-cultural technicians). Due to the obtained results it was only consider 5 micro-environments: 1) bedroom; 2) living-room; 3) canteen; 4) outdoor, and 5) others indoor micro-environments. The number size distributions were measured in bedrooms and living-rooms during the occupied period: all night and all day, respectively. The sampling campaign occurred between October and November of 2012, avoiding extreme temperature and humidity. The number size distribution was measured by the Handheld 3016-IAQ – Lighthouse and the equipment was placed at breathing height (±1.5m) in the middle of the indoor micro-environments. In the indoor of ECCs the measuring time ranged from 7 hours to 16 hours. Data reduction and analysis of the recorded size distribution was performed by arithmetic means of the total sampling period, for each studied indoor micro-environment.

An exhaustive explanation can be seen in Chapter 2 (2.2 Material and Methods).

## 3.3.2 Second approach – personal aerosol measurements

Five autonomous elders were randomly selected to participate on the current study. Their ages ranged from 67 to 77 and all of them were retired (Table 3.1). The elders lived in their non-smoking houses, located in the metropolitan area of Lisbon, the capital city of Portugal. This region is located in the west of Portugal, on the Atlantic Ocean coast, being the western most capital in mainland Europe. The metropolitan area of Lisbon has an area of 2870 km<sup>2</sup> and has almost 3 million inhabitants (INE, 2012). Each elder were followed for one set of 2 to 5 consecutive days in order to measure 24-hr personal particles exposure.

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	Gender	Age
Elder 1	Man	77
Elder 2	Woman	76
Elder 3	Woman	68
Elder 4	Man	67
Elder 5	Woman	68

In addition, elders filled in a time-activity diary to record locations where they had been during each sampling period. Thirteen micro-environments were identified throughout the

time-activity diary: bedroom, living-room, kitchen, gymnasium, garage, car, café, supermarket, public transportation, toilette, outdoor, old office, and indoor balcony. The latest two micro-environments (old office and indoor balcony) were assembled and defined as "Other indoors".

The sampling campaign occurred between March and April of 2014, avoiding extreme temperature and humidity. Five different particle size bins (0.3-1; 1-2.5; 2.5-4; 4-10; >10  $\mu$ m) were measured by the DustTrack aerosol monitor (model 8533, TSI, USA) and the equipment was placed, in a back-bag, as much as possible near the breathing zone (height  $\pm$  150 cm). The elders wear the back-bag during the day and at sleeping time the bag-back was placed near the elders' bed at breathing zone.

In order to assure the comparability of the PM concentration obtained from the DustTrack, an inter-comparison was performed between it and sampled with Gent Sampler and analysed by gravimetry. Both equipments were placed side-by-side. Gent sampler operated at a flow rate of  $16.5~\text{m}^3.\text{h}^{-1}$  during 5 consecutive days (sampling time average = 8.2~hours). The filters were weighted using a Mettler® Toledo balance with  $0.1~\mu g$  readability, placed in a controlled clean room (class 10,000) at a temperature of  $20\pm1^\circ\text{C}$  and a relative humidity of  $50\pm5\%$ , before and after sampling and the mass was obtained as the average of three measurements, when observed variations were less than  $5~\mu g$ . The results are presented in Figure 4.1 and showed that DustTrack overestimate the levels in comparison to gravimetric method, but presents high correlations when compared to gravimetric samplers for different diameters ( $r^2=0.90~\text{and}~r^2=0.94~\text{for PM10}$  and PM2.5, respectively). DustTrack presents the same behaviour as Lighthouse in Chapter 2.3.5.1.

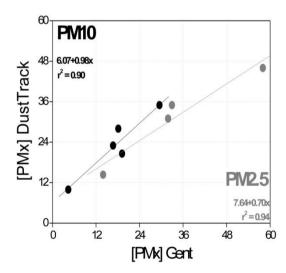


Figure 3.1 – Relation between PM concentration obtained by DustTrack and Gent Sampler (values in  $\mu g.m^{-3}$ ).

Data reduction and analysis of the recorded size distribution was performed by arithmetic means of the total sampling period, for each elder and studied micro-environments.

## 3.4 Modelled data

Deposition of inhaled airborne particles in the respiratory tract is related to both the physical properties of the particles and the anatomical and physiological characteristics of respiration. Exposure characterization and estimation of PM intake are equally important for the assessment of their potential health impacts. Herein we adopt a modelling approach in order to combine these pieces of information.

Particle transport and deposition within the regions of the HRT are determined using a numerical model based on an Eulerian approach describing the air flow and aerosol dynamics in the respiratory tract. The model predicts the temporal variation of the number concentration and the regional deposition of the inhaled particles during a breathing cycle by solving the aerosol general dynamic equation (GDE):

temporal variation convection diffusion deposition
$$\frac{\partial}{\partial t}(A_{t} n_{i}) = -\frac{\partial}{\partial x}(A_{A} u n_{i}) + \frac{\partial}{\partial x}(A_{t} D_{i} \frac{\partial n_{i}}{\partial x}) - U_{d_{i}} \Gamma n_{i}$$

$$+ \left(\frac{\partial}{\partial t}(A_{t} n_{i})\right)_{\text{growth}} + \left(\frac{\partial}{\partial t}(A_{t} n_{i})\right)_{\text{coagulation}} (3.1)$$

where t the time,  $n_i$  the particle number concentration in section i of the size distribution, u the fluid velocity,  $D_i$  the diffusion coefficient of particles with size i,  $A_t$  and  $A_A$  the time dependent and constant cross-section of all airducts, respectively, at distance x from the respiratory system entrance,  $\Gamma$  the circumference of airducts and  $U_{di}$  the particle deposition velocity. The GDE is considered in a one dimensional form along the flow direction and describes the different processes (convection, axial diffusion, deposition, condensational growth, coagulation) acting simultaneously on the inhaled particulate matter. The description of the above deposition mechanisms is based on standard theory for the respective aerosol processes, avoiding the use of empirical correlations. The respiratory tract consists of the alveolar-interstitial, thoracic (lung) and the extrathoracic regions (Figure 3.2). The thoracic region of the respiratory tract is described with the help of the classical morphometric model "A" by Weibel (Weibel, 1963). The volume of the alveolated section of the lung is left to vary with time to accommodate effects due to breathing dynamics. A simplified morphological scheme that consists of sequential cylindrical airways describes the extrathoracic region through the mouth pathway. The air

velocity along the airways of the respiratory tract is determined by solving the equation of continuity.

The model has been extensively validated against a large body of experimental and numerical respiratory data, for both inert and hygroscopic aerosols, and the predictions of the empirical model that is used by the International Commission on Radiological Protection (ICRP). A detailed description of the model, its validation and application potential can be found in literature (Mitsakou et al., 2007a,b; Mitsakou et al., 2005).

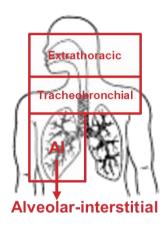


Figure 3.2 – Scheme that represents the different regions of the human lungs.

It was assumed that the state of elders in the bedroom was sleeping, in the living room and indoor balcony was sitting awake, in the gym was heavy exercise and for the rest activities was light exercise. The physiological parameters used for the calculations are shown in Table 3.2 (ICRP, 1994). In addition, coagulation was neglected because it was thought irrelevant for the particle population under study; coagulation is one mechanism that affects mostly the ultra-fine particles (UFPs), and in fact its effect is stronger for the smaller UFPs (Rim et al., 2012). In all cases the values corresponded to a healthy, adult, Caucasian male or female subject.

Table 3.2 - Reference respiratory values at different levels of activity (ICRP, 1994). (FRC: Functional Residual Capacity,  $V_T$ : Tidal Volume,  $f_R$ : Respiration Frequency)

MALE								
Activity	FRC [10 <sup>-3</sup> m <sup>3</sup> ]	$V_{T} [10^{-3} \text{ m}^{3}]$	<b>f</b> <sub>R</sub> [min <sup>-1</sup> ]					
Resting (sleeping)		0.625	12					
Sitting Awake	3.3	0.750	12					
Light Exercise		1.25	20					
	FEMALE							
Activity	FRC [10 <sup>-3</sup> m <sup>3</sup> ]	$V_{T} [10^{-3} \text{ m}^{3}]$	$f_{\rm R}$ [min <sup>-1</sup> ]					
Resting (sleeping)		0.444	12					
Sitting Awake	2.68	0.464	14					
Light Exercise	2.00	0.992	21					
Heavy Exercise		1.364	33					

# 3.5 Measured parameters

## 3.5.1 PMx and time-occupancy data in institutionalized elders

All the results related to PM levels measured in the 10 ECC were already presented in Chapter 2 (2.4.1 Elderly Daily Pattern and 2.4.3 CO, O<sub>3</sub>, VOC, CH<sub>2</sub>O, PMx concentrations and Comfort Parameters in 10 ECCs). Here, it is presented a brief discussion to introduce the next sub-chapter.

Particle mass concentrations in difference size per micro-environment are presented in Table 3.3. In average, there were no statistically differences between each ECC (p-value > 0.05) for all particles sizes. Results showed that in average PM concentrations in living-rooms were significantly higher than in bedrooms, except for ECC 7 (p = 0.79). Living-rooms of ECC 4, ECC 5, ECC 7 and ECC 10 presented the highest PM10 average concentration (44, 43, 41 and 47  $\mu$ g.m<sup>-3</sup>, respectively). For ECC 4 and ECC 5, PM10 maximum values were 860 and 347  $\mu$ g.m<sup>-3</sup>, respectively. These concentrations exceeded the limit value of 50  $\mu$ g.m<sup>-3</sup> defined by the Portuguese legislation (Portaria 353-A/2013). ECC 7 and ECC 10 did not exceed the limit value. The high PM10 concentration measured in ECC 7 can be explained by the high outdoor PM10 concentration (71  $\mu$ g.m<sup>-3</sup>). The bedrooms of ECC 7 and ECC 10 presented a PM2.5 average concentration of 21  $\mu$ g.m<sup>-3</sup> and 34  $\mu$ g.m<sup>-3</sup>, respectively.

The average particle concentrations measured in this work were similar to those found in a study developed with elders living in Amsterdam and Helsinki that presented PM2.5 average concentrations of  $16~\mu g.m^{-3}$  and  $11~\mu g.m^{-3}$ , respectively (Lanki et al., 2007). In UK houses PM10, PM2.5 and PM1 concentrations (13, 6 and 3  $\mu g.m^{-3}$ , respectively) were lower comparing with current work (Nasir & Colbeck, 2013). In an Italy study, the average

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PM2.5, PM5 and PM10 concentration presented lower results comparing with current work. Nevertheless, the same study showed higher concentrations in lower particle sizes 18 and 9.2 μg.m<sup>-3</sup> (PM0.5 and PM1, respectively). This fact could be due to the strong association between indoor PM0.5 and the number of cigarettes smoked (Urso et al., 2015), since previously studies had already demonstrated that environmental tobacco smoke is the most significant source of indoor-generated fine particles (Chen & Zhao, 2011). PM2.5 and PM10 average concentration measured in several houses demonstrated a similarity of results: 7.9 μg.m<sup>-3</sup> and 17 μg.m<sup>-3</sup> (Jones et al., 2000); 9.1 μg.m<sup>-3</sup> and 23 μg.m<sup>-3</sup> (Lawson et al., 2011); 7.3 μg.m<sup>-3</sup> and 22 μg.m<sup>-3</sup> (Molloy et al., 2012). However, it is possible to find studies with higher particles concentrations, such as the one developed by Chao (2002) where PM2.5 and PM10 average concentration of 45 μg.m<sup>-3</sup> and 63 μg.m<sup>-3</sup> were measured, respectively. Those values could be explained by the existence of different sources: smoking, cooking and burning incense (Urso et al., 2015; Chao & Wong, 2002).

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Table 3.3 - Measured particle mass concentration per ECC. Results are presented in μg.m<sup>-3</sup> (AVG= Average and STD= Standard Deviation).

				Parti	icle Mass (	Concentra	tion (µg.m	-3)				
	Bedroom											
	ECC1	ECC2	ECC3	ECC4	ECC5	ECC6	ECC7	ECC8	ECC9	ECC10	AVG	STD
PM0.5	2.2	1.4	1.3	2.0	3.3	1.1	7.9	1.1	1.8	19.3	4.1	5.7
PM1	3.1	2.5	2.2	3.1	4.7	1.7	14	2.7	2.7	28	6.5	8.4
PM2.5	4.6	4.8	4.2	5.9	9.2	2.8	21	8.9	4.7	34	10	10
PM5	8.1	11	11	16	15	3.9	29	12	7.0	40	15	11
PM10	10	16	17	24	29	6.2	41	16	11	47	22	13
						Living-	room					
	ECC1	ECC2	ECC3	ECC4	ECC5	ECC6	ECC7	ECC8	ECC9	ECC10	AVG	STD
PM0.5	0.22	2.5	4.7	2.6	2.6	4.9	2.7	0.72	1.9	2.5	2.5	1.5
PM1	1.0	4.2	6.9	4.3	4.6	7.5	5.1	2.3	2.7	4.5	4.3	2.0
PM2.5	4.2	8.2	10	10	11	11	16	9.4	4.2	12	9.6	3.5
PM5	14	23	21	28	19	15	25	14	6.2	19	18	6.5
PM10	19	34	29	43	43	27	35	19	11	31	29	10

Comparing the living-room with the bedroom it is possible to observe that the coarse fraction was dominated in living-room whereas fine fraction was dominated in bedroom. This fact indicates the importance of particles re-suspension in living-room.

As it was said before, elderly are considered a susceptible population such as children. By this reason and because studies performed in ECCs are rare, results from the current work were compared with the ones developed in schools. The largest difference between ECCs and schools is the behaviour of the population - most of the elders have reduced movement capacities whereas children are in constant activity promoting a higher re-suspension of dust. Studies developed in Lisbon schools showed that children were exposed to a PM2.5 concentration of 10 μg.m<sup>-3</sup> and PM10 concentrations varied from 30 μg.m<sup>-3</sup> to 146 μg.m<sup>-3</sup> (Canha, et al, 2012a; Almeida, et al., 2011). Another study developed in three different Polish schools referred that average particles concentrations varied between 94 μg.m<sup>-3</sup> and 191 μg.m<sup>-3</sup> for PM10 and 45 μg.m<sup>-3</sup> and 119 μg.m<sup>-3</sup> for PM2.5 (Polidnik, 2013).

Several studies have already evaluated the daily time pattern of people from different countries (Fisher & Robinson, 2011; Eurostat, 2006, 2003). However, these studies either excluded the old people or studied simultaneously all age groups, since young children to elderly.

Table 3.4 shows the time spent by the elders in each micro-environment. Due to the lack of differences between weekdays and weekends, the results are presented for typical 24 hours. The micro-environment referred as "Others" is defined as other indoor micro-environments inside and/or outside the elderly care centres, such as family houses, restaurants or coffees.

Table 3.4 - Time-budget data for all 384 voluntaries in 24 h per studied site (values in percentage).

=	Bedroom	Living-room	Canteen	Outdoor	Others
ECC 1	52	22	19	3	4
ECC 2	62	22	13	2	1
ECC 3	76	15	5	0	3
ECC 4	64	20	9	1	7
ECC 5	57	22	14	7	0
ECC 6	50	42	0	8	0
ECC 7	46	48	0	4	3
ECC 8	55	30	7	8	0
ECC 9	54	46	0	0	0
ECC 10	56	32	11	1	0
Average	57	30	8	3	2

Old people in ECCs spent the majority of their time inside bedrooms (57%) and living-rooms (30%). Due to this evidence, these two micro-environments were chosen to perform a detailed IAQ characterization. In ECC 3 people spent more time in bedrooms due to the high number of bedridden (13%) in this institution. On the other hand, in ECC 1 and ECC 7 all the elders are daily lifted and placed in the living-rooms, which can explain the less percentage of time spent inside the bedrooms. In all ECC the same pattern was observed: from 8:00 to 20:00 the majority of the elders were in living-room, moving to the canteen at the meal times and to the bedrooms at 21:00.

## 3.5.2 PMx and time-occupancy data in autonomous elders

Regarding the personal exposure to PM by micro-environments, the kitchen was the micro-environment with highest concentration in all particle sizes (41.2; 41.8; 42.9 and 46.6  $\mu$ g.m<sup>3</sup> for size bins 0.3-1, 1-2.5, 2.5-4 and 4-10  $\mu$ m, respectively), as shown in Figure 3.3. Statistically, the indoor PM concentrations were not different comparing to outdoor results (p > 0.05).

Even being the bedroom the micro-environment with lowest PM levels, with average concentrations of 9.6, 9.9, 10.5 and 12.1  $\mu$ g.m<sup>-3</sup> size bins 0.3-1, 1-2.5, 2.5-4 and 4-10  $\mu$ m, respectively, the results were not significantly statistic different between each micro-environment. This study also showed that the various micro-environments have different levels of contribution to daily personal exposure, which not only results from the actual particles concentration in each site but also on the amount of time that people spend there.

In the right side of Figure 3.3, it is presented the indicative personal exposure time-series, in order to identify the existence of peaks. According to the results, a decrease on particles concentration during the night period can be observed, as expected, since particles commonly deposit in the absence of movement (no re-suspension). The first peak that occurred between 7:00 and 9:00 corresponds to the uprising and cooking period (breakfast in the kitchen). The other peaks are associated with the public transportations – period of commuting between home and other indoors. There were not significant differences between particles sizes, however it is possible to observe that PM1 is the principal contributor to the exposure (Figure 3.3 right).

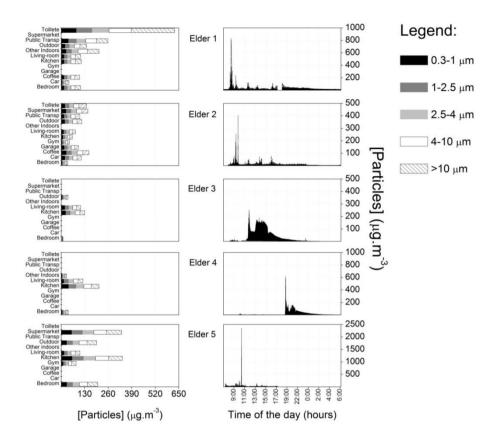


Figure 3.3 – Particles concentration in each micro-environment (left) and temporal variation of particles concentration (right) per elder. Results are presented in µg m<sup>-3</sup>.

The personal daily exposure to PM discriminated by granulometry is present in Table 3.5. The results for total PM concentration varied from 14.2  $\mu g.m^{-3}$  for Elder 2 to 44.2  $\mu g.m^{-3}$  for Elder 5. An interesting finding of the experimental campaign was that all the elders were predominantly exposed to PM1; from 74% for Elder 5 to 100% for Elder 1. Elder 4 was an exception because only half of his exposure could be attributed to PM1, had being also exposed to a considerable amount of coarse particles (>PM4).

In average, Elder 5 was subjected to the highest personal daily exposure in all particles sizes. The 24-hr maximum PM concentration was observed during a cooking period. In general, Elders 1, 2 and 3 presented lower personal daily exposure, not only comparing with the other 2 studied elders but also with the findings of the Rojas-Bracho et al. (2004) study. Additionally, comparing the present results with a study developed in Beijing, with college students, that presented a PM<sub>2.5</sub> daily exposure of 163 µg.m<sup>-3</sup>, it is evident that

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elders were exposed to considerably lower concentrations of PM during their daily activity (Zhu et al., 2010). Regardless the low concentrations measured in this study, it is well-known that PM enhances adverse health effects and it is unclear whether a threshold concentration exists for PM below which no effects on health are likely. Moreover, it should not be forgotten that elderly population is more susceptible to air pollutants.

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Table 3.5 - Personal dail	y exposure to particle	s. Results presente	ed in ug.m°.

	PM1 (STD)	PM2.5 (STD)	PM4 (STD)	PM10 (STD)	TOTAL (STD)
Elder 1	18 ( 3.6)	$18 \pm 3.6$	$18 \pm 3.6$	$18 \pm 3.6$	$18 \pm 3.6$
Elder 2	14 (14)	$14 \pm 14$	$14 \pm 14$	$14 \pm 14$	$14 \pm 14$
Elder 3	13 (4.3)	$13 \pm 4.2$	$13 \pm 4.1$	$14 \pm 3.6$	$15 \pm 3.1$
Elder 4	20 (4.2)	$21 \pm 4.1$	$23 \pm 4.0$	$28 \pm 3.0$	$38 \pm 5.1$
Elder 5	33 (22)	$33 \pm 22$	$35 \pm 22$	$40 \pm 21$	$44 \pm 22$

Figure 3.4 shows the micro-environments contribution to each elders' exposure. The greatest differences were among the elders, due to the differences in their daily routine. Living-room and kitchen were, in average, the two micro-environments that highly contributed to the elders' daily exposure to particles, even when they did not spend the majority of their daily time on those micro-environments. The results were in accordance with a previously study developed with institutionalized elders where the living-room was the micro-environment that highly contribute to their daily exposure to particles (Almeida-Silva et al., 2015). The micro-environment defined as "Other Indoors" presented an important contribution to the daily exposure of the Elder 4 (almost 50%). This can be explained by the fact that this elder spent a great part of their daily time in an indoor balcony, doing some handwork (with wood). Figure 3.4 also showed that the bedroom did not contribute more than 25% to the elders' daily exposure, even when they spent in average 33% of their daily time on that micro-environment.

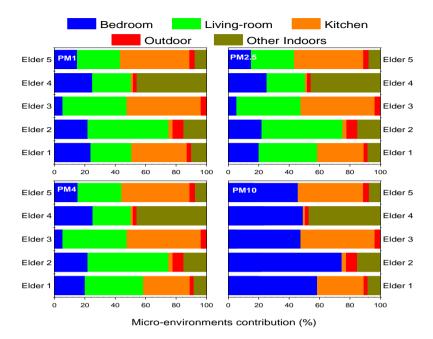


Figure 3.4 – Contribution of micro-environments to elderly daily exposure to particles. Values are presented in %.

Figure 3.5 shows the distribution of time spent by the volunteers in various micro-environments. The results are presented for a typical day (24 hours), being the average exposure over the whole sampling period. Locations were classified in four micro-environments: living-room, bedroom, kitchen, outdoor, and other indoors. In average, the studied elders spent 95% of their time in indoor environments, being 82% of the time spent inside their homes. Women spent, in average, 42% and 18% of their time in living-room and kitchen, respectively, while men spent the majority of their time in bedroom (36%) and other indoor micro-environments, such as the old office, café, public transportation and/or indoor balcony (25%).

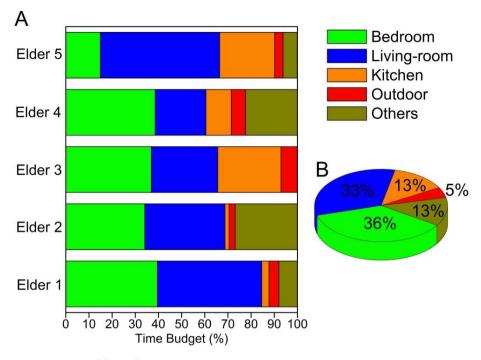


Figure 3.5 – Elders' time-activity pattern. Results are presented in percentage (%).

Although several studies have already evaluated the daily time pattern of people from different countries (Fisher and Robinson, 2011; Eurostat, 2006, 2003), these studies either excluded old people or studied simultaneously all age groups, from young children to elders. A study developed in Italy showed that elders spent 70-83% of their time inside buildings (Simoni et al., 2003). In addition, the National Human Activity Pattern Survey (NHAPS) developed in USA refers that the American elders spent 87% of their time indoors (Klepeis et al., 2001). The results of the present study are in line with both the aforementioned works.

# 3.6 Modelled parameters

The HRT particles transport and deposition model was used in order to calculate the daily burden of PM in the respiratory tract of the elders. It should be noted that in the present study, hygroscopic growth was not considered in the calculations due to a lack of information regarding the nature of the particulate matter.

## 3.6.1 Internal dose of particles – institutionalized elders

The ratio between the particles that were deposited into the HRT and the particles that were inhaled, i.e. the deposition fraction DF, discriminated by particles number and regions of deposition in the HRT are shown in Figure 3.6 for both genders in the bedroom and the living-room. The presented DFs were calculated using the average measured particles concentrations over all ECCs, as shown in Table 3.6. In all cases, total DF was higher in male than female for the same activity level. However, the DF in the alveolar-interstitial (AI) region was always higher for the females. This means that the particles penetrated deeper in the lungs of women, whereas they were better filtered in the extrathoracic (ET) region of men. Moreover, for both genders, DF increased with activity level, because of the more frequent breathing and the larger volumes of air consumed.

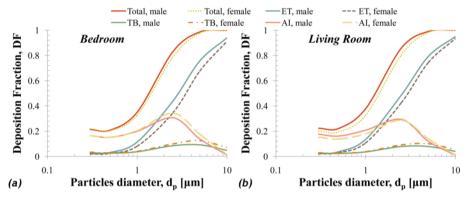


Figure 3.6 – Deposition fraction, DF, per particle diameter in each region of the HRT for males and females; (a) Bedroom, and (b) Living Room (ET: extrathoracic, TB: Tracheobronchial, AI: alveolar-interstitial).

The average, over both gender and ECCs, daily dose of deposited particles in the whole HRT and its different regions, i.e. in the extrathoracic (ET), the tracheobronchial (TB) and the alveolar-interstitial (AI), was given in Table 3.5. It is shown that the daily dose was more than 91% attributed to the particles sized between 0.3-1µm. In addition, in Figure 3.7, the distribution of the daily dose in the different regions of the HRT was given per particle diameter and for the total particle population. Almost 70% of the deposited particles ended up in the deeper parts of the lungs, the AI region.

Table 3.6 - Total number of deposited particles in the HRT regions per day (average for
both genders). (ET: extrathoracic, TB: Tracheobronchial, AI: alveolar-interstitial)

Daily Dose [#]								
dp [μm]	TOTAL	ET	TB	AI				
0.3-0.5	234	22	31	181				
0.5-1	92	15	11	67				
1-2.5	46	16	4.8	25				
2.5-5	32	18	3.2	11				
5-10	2.7	2.1	0.24	0.35				
>10	1.4	1.3	0.07	0.01				
TOTAL	409	74	50	284				

It was also interesting to see the contribution of each micro-environment in the total daily dose of particles in the whole HRT. It was found that the living-room was by far the micro-environment that contributed more to the daily dose in all ECCs, regardless of the considerably less time spent there comparing with the bedroom, in most ECCs (Figure 3.8).

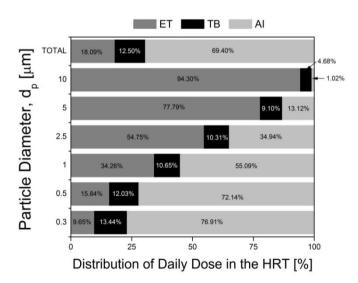


Figure 3.7 – Distribution of deposited particles in the HRT based on the total daily dose for each particle diameter and the total particles population.

In average over all ECCs and both genders, the dose from the living room was related with all particle sizes under study (Figure 3.8). On the contrary, the most significant contribution of the bedroom was on the dose from particles of diameters less than  $0.5\mu m$ .

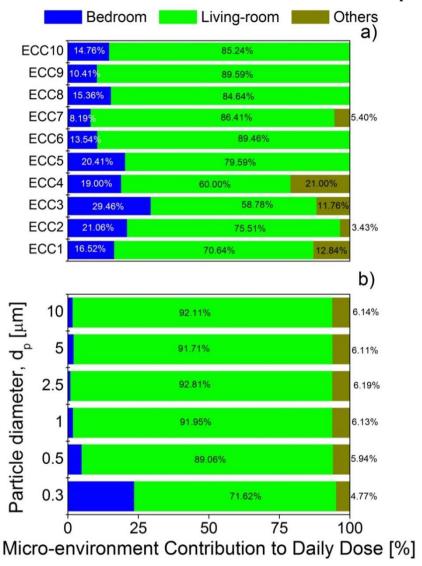


Figure 3.8 – Variability of micro-environment contribution to the total HRT daily dose between the Elderly Care Centres (a) and particle diameter (b) (average for both genders).

# ${\bf 3.6.2\ Internal\ dose\ of\ particles-autonomous\ elders}$

In Table 3.7 the daily dose of PM in the HRT is given for each elder, as well as the percentage of this dose in each part of the respiratory system.

Table 3.7 - Daily dose (in  $\mu$ g) per elder and its distribution in the different regions of the HRT. (ET: extrathoracic, TB: tracheobronchial, AI: alveolar-interstitial).

	Daily	Distribution in the HRT					
	Dose [µg]	ET	TB	AI			
Elder 1	392	75%	6.5%	19%			
Elder 2	90.0	58%	9.1%	33%			
Elder 3	139	53%	9.8%	37%			
Elder 4	156	52%	8.8%	39%			
Elder 5	488	68%	7.9%	24%			

The daily dose depends on the unique mixture of activities performed by each individual and the physical excitement level, the exposure to PM and the time spend on each of these activities. Thus, marked differences were observed in both the total daily dose and its distribution in the HRT between the individuals, e.g. the daily dose ranged from almost 90  $\mu$ g for Elder 2 to almost 500  $\mu$ g for Elder 5. For all individuals, more than half of the PM deposited in the extrathoracic region, varying from ~52% (Elder 4) to ~75% (Elder 1), whereas less than 10% deposited in the tracheobronchial. The percentage of PM that deposited in the alveolar-interstitial region of the lungs ranges considerably between the elders, from ~19% (Elder 1) to ~39% (Elder 4), an indication of the quantity of small particles that each one was exposed to.

In fact, for Elders 3 and 4 that spent the vast majority of their time indoors (93% and 94%, respectively), their daily dose was attributed almost exclusively to sub-micron particles (Figure 3.9). For Elders 1, 2 and 5 considerable contribution of particles greater than 4  $\mu$ m was also observed. These elders were exposed to such particles especially in the kitchen and other indoor environments (Table 3.8).

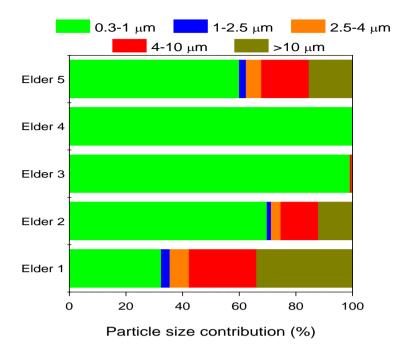


Figure 3.9 – Particle size contribution to daily dose.

Finally, in Figure 3.10 the contribution of the micro-environment to the daily dose is shown for each elder. It is clear that the kitchen was a major contributor to the daily dose for the individuals that spent considerable part of their time in there, i.e. Elders 3, 4 and 5. Moreover, the outdoor activities contribute around 5-6% to the total daily dose, with the exception of Elder 2, for which outdoor activities correspond to ~22% of her daily dose. In all cases, it is obvious that the activities taking place indoors were responsible for the vast majority of the daily PM dose in the respiratory tract of all individuals.

# **Chapter III**

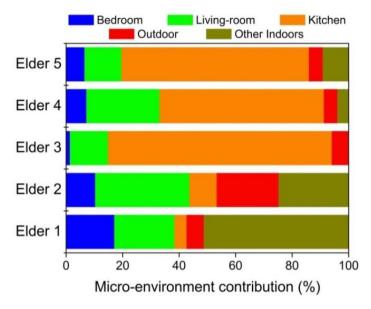


Figure 3.10 – Micro-environment contribution to the elder's daily dose.

Table 3.8 - Daily dose in the HRT and its regions per microenvironment, particles size range and elder. Results are presented in  $\mu g$ . (ET: extrathoracic, TB: Tracheobronchial, AI: Alveolar-Interstitial).

	-								D.	III V D	OSE /µg	,,								
		Elder	1 (M)			Flde	r 2(F)		DA		υ 3 (F)	<i>i</i>		Elder 4	(M)			Elder	5 (F)	
Bedroom		Liuci	1 (111)			Liuc	1 2(1)			Little	13(1)			JIGCI 4	(111)			Liuci	J (I)	
D <sub>p</sub> [μm]	Total	ET	TB	ΑI	Total	ET	ТВ	AI	Total	ET	ТВ	AI	Total	ET	ТВ	ΑI	Total	ET	TB	ΑI
0.3-1	22.00	7.1	2.3	12	6.9	1.7	0.86	4.3	1.9	0.48	0.24	1.2	11	3.7	1.2	6.5	9.7	2.4	1.2	6.0
1-2.5	2.50	1.1	0.26	1.1	0.18	0.065	0.022	0.093	0.0025	0.00	0.00	0.00	0.00	0.00	0.00		0.54	0.19	0.07	0.28
2.5-4	4.50	2.7	0.46	1.3	0.31	0.16	0.041	0.11	0.0073	0.00	0.00	0.00	0.00	0.00	0.00	0.00	2.0	1.0	0.26	0.71
4-10	12.00	10	0.86	0.60	0.89	0.73	0.10	0.062	0.030	0.024	0.00	0.00	0.00	0.00	0.00	0.00	7.6	6.2	0.81	0.53
>10	27.00	25	1.3	0.30	1.0	0.92	0.072	0.015	0.017	0.016	0.00	0.00	0.00	0.00	0.00	0.00	12	11	0.88	0.18
Total	67	46	5.2	16	9.3	3.6	1.1	4.6	2.0	0.52	0.24	1.2	11	3.7	1.2	6.5	32	21	3.2	7.7
Living Roo	om																			
$D_p [\mu m]$	Total	ET	TB	AI	Total	ET	TB	AI	Total	ET	TB	AI	Total	ET	TB	ΑI	Total	ET	TB	AI
0.3-1	29	10	2.7	16	20	6.2	2.4	12	19	5.7	2.2	11	40	14	3.8	22	27	8.4	3.3	16
1-2.5	1.9	0.90	0.17	0.79	0.35	0.15	0.04	0.16	0.005	0.00	0.00	0.00	0.00	0.00	0.00	0.00	1.6	0.70	0.19	0.75
2.5-4	5.1	3.3	0.46	1.4	1.1	0.63	0.13	0.33	0.007	0.00	0.00	0.00	0.00	0.00	0.00	0.00	5.3	3.0	0.61	1.6
4-10	23	20	1.4	0.99	4.7	4.0	0.41	0.27	0.053	0.045	0.00	0.00	0.00	0.00	0.00	0.00	17	14	1.5	0.96
>10	25	23	0.98	0.22	3.4	3.2	0.20	0.04	0.040	0.037	0.00	0.00	0.00	0.00	0.00	0.00	13	12	0.73	0.16
Total	83	58	5.8	19	30	14	3.2	12	19	5.8	2.2	11	40	14	3.8	22	64	38	6.3	19
Kitchen																				
$D_p [\mu m]$	Total	ET	TB	ΑI	Total	ET	TB	ΑI	Total	ET	TB	ΑI	Total	ET	TB	ΑI	Total	ET	TB	ΑI
0.3-1	6.1	3.8	0.51	1.8	5.2	3.0	0.50	1.8	109	62	10	37	91	56	7.6	27	224	126	21	76
1-2.5	0.45	0.32	0.036	0.088	0.18	0.12	0.017	0.041	0.090	0.061	0.008	0.021	0.02	0.01	0.00	0.00	8.3	5.6	0.77	1.9
2.5-4	0.94	0.80	0.058	0.082	0.30	0.24	0.022	0.032	0.13	0.10	0.010	0.014	0.00	0.00	0.00	0.00	17	14	1.2	1.8
4-10	3.3		0.087	0.026	1.1	1.0	0.037	0.011	0.50	0.48	0.017	0.00	0.00	0.00	0.00	0.00	46	44	1.6	0.46
>10	6.2	6.1	0.089	0.0062	1.9	1.8	0.034	0.0023	0.28	0.28	0.005	0.00	0.00	0.00	0.00	0.00	29	29	0.54	0.04
Total	17	14	0.78	2.0	8.7	6.2	0.60	1.9	110	63	10	37	91	56	7.6	27	324	219	25	81
Outdoor		****	-			****	-								-			****	-	
D <sub>p</sub> [μm]		ET	TB	AI	Total	ET	TB	AI	Total	ET	TB	AI	Total	ET	TB	AI	Total	ET	TB	AI
0.3-1	12	7.2	0.97	3.5	14	7.7	1.3	4.7	7.9	4.5	0.75	2.7	7.5	4.6	0.63	2.2	11	6.0	0.99	3.6
1-2.5 2.5-4	1.0	0.75	0.083	0.20	0.28	0.19	0.026	0.063	0.030	0.021	0.0028	0.0069	0.00	0.00	0.00		0.50	0.34	0.046	0.11
4-10	4.6	4.5	0.11	0.10	2.6	2.5	0.030	0.080	0.038		0.0029	0.0041	0.00	0.00	0.00	0.00	5.3	5.1	0.088	0.13
>10	4.6	4.5	0.12	0.037	2.3	2.3	0.088	0.028	0.155	0.127	0.0044	0.0013	0.00	0.00	0.00	0.00	6.4	6.3		0.0078
Total	24	19	1.4	3.9	20	13	1.5	4.9	8.2	4.7	0.76	2.7	7.5	4.6	0.63	2.2	24	19	1.4	3.9
Other Indo		17	1.7	3.7	20	13	1.3	4.7	0.2	7./	0.70	2.7	7.3	7.0	0.03	2.2	24	1)	1.7	3.7
D <sub>p</sub> [μm]		ET	ТВ	AI	Total	ET	тв	AI	Total	ET	ТВ	AI	Total	ET	ТВ	ΑI	Total	ET	ТВ	ΑI
0.3-1	58	28	5.2	25	16	9.3	1.6	5.5	0.00	0.00	0.00	0.00	6.1	2.2	0.58	3.3	22	13	2.1	7.1
1-2.5	6.2	3.9	0.53	1.8	0.30	0.21		0.060	0.00	0.00	0.00	0.00	0.00	0.00	0.00		0.62	0.43	0.06	0.13
2.5-4	14	10	1.1	2.7	0.65	0.53		0.068	0.00	0.00	0.00	0.00	0.00	0.00	0.00		1.6	1.3	0.11	0.15
4-10	51	47	2.8	1.9	2.5	2.4		0.029	0.00	0.00	0.00	0.00	0.00	0.00	0.00		6.6	6.3	0.21	0.06
>10	70	67	2.6	0.58	2.2	2.2	0.043	0.0042	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	14	14	0.24	0.02
Total	200	156	12.3	32.1	22.1	14.6	1.8	5.7	0.00	0.00	0.00	0.00	6.1	2.2	0.58	3.3	44	34	2.7	7.5
TOTAL																				
D <sub>p</sub> [μm]	Total	ET	TB	AI	Total	ET	TB	AI	Total	ET	TB	AI	Total	ET	TB	ΑI	Total	ET	TB	ΑI
0.3-1	127	57	12	59	63	28	6.6	28	138	72	14	52	156	81	14	61	293	156	29	109
1-2.5	12	7.0	1.1	4.0	1.3	0.74	0.13	0.42	0.13	0.08	0.012	0.031	0.016	0.01	0.00		12	7.3	1.1	3.2
2.5-4	26	19	2.2	5.6	3.1	2.2	0.29	0.62	0.18	0.14	0.014	0.022	0.00	0.00	0.00		27	20	2.3	4.4
4-10	93	85	5.3	3.6	12	11	0.72	0.40	0.72	0.67	0.029	0.011	0.00	0.00	0.00	0.00	82	76	4.2	2.1
. 10																				0.40
>10	133	126	5.1	1.1	11	10	0.39	0.07	0.39	0.38	0.010	0.0012	0.00	0.00	0.00	0.00	75	72	2.5	0.40

## 3.7 Conclusions

In the present work, PM measurements, in 10 Elderly Care Centers, were combined with a mechanistic numerical model in order to calculate the dose in the different parts of the human respiratory tract for the elders living in these ECCs. Plus, five volunteers carried out a PM sampler in order to calculate not only their personal exposure to particles but also to calculate their deposited dose of particles in HRT.

The time-budget survey, applied to institutionalized elders, showed that elders spent 57% and 30% of their time in bedroom and living-room, respectively, in a total of 97% of their

time in indoor environments, while autonomous elders spent 95% of their time in indoor environments. Results from this work clearly showed that exposure to particles, even at low levels of concentration, results in alterations of the daily dose.

Considering the results provided from the application of first approach, the following were observed:

#### Measured data:

- PM concentrations measured in living-room were significantly higher than in bedroom;
- Coarse and fine PM fractions were associated with living-room and bedroom, respectively;
- High concentration of coarse fraction in living-room were related to resuspension of particles;

#### Modelled data:

- Deposition fraction was higher in males than females for the same activity level. However, the DF in the alveolar-interstitial region was always higher for the females;
- 70% of the deposited particles ended up in the deeper parts of the lungs;
- More than 91% of the daily dose was attributed to particles sized between 0.3-1μm;
- Living-room was the micro-environment that contribute more to the elders' daily dose of particles;
- Bedroom had the highest contribution to the daily dose regarding the particles with diameter lower than 0.5μm.

Considering the results provided from the application of second approach, the following was observed:

#### Measured data:

- The three women spent, in average, more time in living-room and kitchen, while the two men spent the majority of their time in bedroom and others indoor micro-environments;
- The kitchen was the micro-environment with highest concentration in all particle sizes and the bedroom with the lowest;
- During the night period a decrease in the particles concentration was observed (due to its deposition on the floor and other surfaces), and the peaks appearing between 7 am to 9 am were associated with the uprising, breakfast and public transportations;
- The elder who presented the highest personal daily exposure in all particles sizes was the same that presented highest percentage of time spent in the kitchen;

#### Modelled data:

- Deposition fraction was higher in male than female for the same activity level.
   However, the DF in the alveolar-interstitial region was always higher for the females;
- For elders that spent the majority of their time indoors, their daily dose was attributed almost exclusively to sub-micron particles. For elders that spent less time at home, their daily dose had also considerable contribution of particles greater than 4 µm of aerodynamic diameter.

As an overall remark, the present work showed how the measurement of exposure to airborne particles can be related to internal doses in the HRT. Additionally, even with a carefully selection of volunteers, all of them with the same characteristics, the present results shows a relevant heterogeneity due to the great variety of behaviours, microenvironments, etc. The results are important to understand which areas the elders where were most exposed, to suggest effective mitigation strategies and, so, to reduce their exposure to particles and in extension to reduce adverse health effects. Moreover, the proposed methodology will be helpful in identification of health risks for the elders, reviewing air quality standards in the Elderly Care Centers and assessing effective policy interventions.

Finally, the combination of the calculated deposited dose with toxicological data could lead to a realistic dose-response relation for the specific particulate matter.

# CHAPTER IV. SOURCE APPORTIONMENT OF INDOOR PM10 IN ELDERLY CARE CENTER

## 4.1 Abstract

Source contribution to atmospheric particulate matter (PM) has been exhaustively modelled. However, people spend most of their time indoors where this approach is less explored. This evidence worsens considering elders living in Elderly Care Centers, since they are more susceptible. The present study aims to investigate the PM composition and sources influencing elderly exposure. Two 2-weeks sampling campaigns were conducted one during early fall (warm phase) and another throughout the winter (cold phase). PM10 were collected with two TCR-Tecora® samplers that were located in an Elderly Care Center living-room and in the correspondent outdoor. Chemical analysis of the particles was performed by Neutron Activation Analysis for elemental characterization, by Ion Chromatography for the determination of water soluble ions and by a Thermal Optical technique for measurement of organic and elemental carbon. Outdoor PM<sub>10</sub> concentrations were significantly higher during the day than night (p-value <0.05), as well as Ca<sup>2+</sup>, Fe, Sb and Zn. Both indoor and outdoor PM10 average concentration did not exceeded the guidelines and there were no significantly differences between seasons. The contribution of indoor and outdoor sources was assessed by Principal Component Analysis and showed the importance of the highways and the airport located less than 500 m of the Elderly Care Center for both indoor and outdoor air quality.

## 4.2 Introduction

Airborne particulate matter (PM) is of major concern since several epidemiological studies have established associations between human exposure to particles and adverse human health effects (Almeida et al, 2014a; Pope et al., 2011, 2002). More recently, some researchers have investigated the properties of ambient aerosol which are responsible for health effects; whether certain particulate chemical components are more harmful than others (Suh et al., 2011; Zanobetti et al., 2009); and the particle size as an important cause of the site and efficiency of pulmonary deposition (Anderson et al., 2008). Thus, indoor concentrations of PM compounds have been sparsely investigated and epidemiological associations between PM and health outcomes are based predominantly on ambient air measurements. In order to apply the best strategies to mitigate the people exposure to particles and consequently to reduce health effects it is important not only to evaluate the PM properties in indoor environments but also to recognize its emissions sources, since the

focus should be done on intervention. Subsequently, the apportioning of indoor particles is so important.

The indoor air concentrations can be affected by several processes (Figure 4.1). Outdoor PM is the main contributor to the indoor PM concentration, being introduced by infiltration (Chen & Zhao, 2011). Particles can also be tracked indoors, be carried on shoes and clothes. The size of the particles influences the differences on deposition velocity: the coarse and the fine particles have higher and lower deposition rates, respectively (Li et al., 2013). However, the deposition rate is not only size-dependent, there are other influencing factors such as airflow pattern, turbulence level, and properties of indoor surfaces that can affect indoor particle deposition (Chen and Zhao, 2011). Particle penetration factor is another important factor that influence the indoor particles concentration. This factor should be approximately equal to 1 if the building is naturally ventilated by opening windows. Considering the size of the particles, larger particles are mostly affected by gravitational settling while ultrafine particle penetration is contrasted by Brownian diffusion (Urso et al., 2015). Some PM will be deposited on the floor and/or furniture and, depending on indoor activities, they could re-suspend.

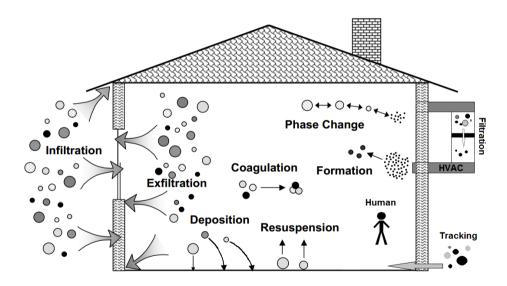


Figure 4.1 – Particle transport and removal processes in the indoor environment [Adapted from Gundel and Destaillats (2013). Chapter 6, Aerosol handbook: measurement, dosimetry and health effects 2<sup>nd</sup> edition].

As far as we know, source apportionment of indoor PM10 and its components has been poorly examined, leading to a lack of indoor sources identification and, consequently, to a deficits mitigation actions.

Considering the scale of the problem, the need of increasing the knowledge seems urgent. During the last few decades the source apportionment studies have been done in outdoor environments, which explain the existence of well-known outdoor emissions sources and fingerprints (Almeida et al., 2013b; Ciaparra et al, 2009). However, the composition of indoor PM is very complex, with similarities but also differences to outdoor aerosols, leading to a totally different type of sources. Some of the indoor coarse particles have been identified to be coming from (e.g.): sweeping, dusting, human movement (walking, dancing, children playing), re-suspension from clothes, re-suspension from carpets, smoking, cosmetics and cooking (Saraga et al., 2010 a,b). Nevertheless, the identification of indoor emission sources is still scarce.

Source apportionment of indoor particles and its components has been poorly examined, leading to a lack of indoor sources identification and, consequently, to a deficit mitigation actions. Therefore, the characterization of indoor particles and the assessment of their sources constitute a major challenges to be addressed in order to fully understand and quantify the magnitude of both individual and population exposure to airborne particles. The present study aims to conduct a comprehensive characterization of the chemical properties of PM10 in indoor environments of an Elderly Care Center (ECC) and to use source apportionment tools, commonly used in outdoor environments, to identify emission sources. This evaluation will be useful to develop appropriate control strategies to minimize the adverse health effects of institutionalized elders.

## 4.3 Materials and methods

# 4.3.1 Sampling site and Elderly Care Center description

The current work was carried out in the metropolitan area of Lisbon, the capital city of Portugal. This region is located in the west of Portugal, on the Atlantic Ocean coast, being the westernmost capital in mainland Europe. The metropolitan area of Lisbon has an area of 2870 km² and has almost 3 million inhabitants. Loures is one of the 18 regions that belongs to the metropolitan area of Lisbon, having around 205 thousands inhabitants in 160 km² of area (INE, 2012). A previously study, published elsewhere, has assessed the indoor air quality in10 Elderly Care Centers (Almeida-Silva et al., 2015, 2014a). According to the obtained results, it was assumed that the ECC selected for the current study was the one that presented more potential issues.

The studied ECC is located in an urban area in Loures and is situated less than 500 m from the international airport of Lisbon (Figure 4.2). Its localization is also near two important highways – CRIL and A1 – which had an annual average daily traffic around 114 and 91 thousands cars in 2009, respectively (INIR, 2010). The ECC has capacity for 69 elders and can be described as a villa with 3 floors and with natural ventilation. The living-room of the ECC was chosen to be the sampling site indoors, due to two main reasons provided from a previously study (Almeida-Silva et al., 2014a): 1) elders spend in average 57% and 30% of

## **Chapter IV**

their time in bedrooms and living-rooms, respectively; 2) the PM concentration in living-room were significantly higher than in bedrooms. The pavement and the windows were made by tile and double glass aluminium, respectively. The selected ECC has typical routines that differentiate it from a normal villa. The movement indoors of the elders and supporters and the cleaning routines are just a few characteristics that make this building different from a normal villa. The kitchen and the canteen are located at a different area of the building, with no direct connection with the living-room.

Air exchange rates ( $\alpha$ ) [renovation per hour ( $h^{-1}$ )] and ventilation rates ( $Q_1$ ) [air litters per second per person (lps.person<sup>-1</sup>)] were calculated for all sites using the build-up method based on  $CO_2$  concentrations as a tracer gas (Canha et al., 2013; Hänninen, 2013). Using  $CO_2$  as tracer gas represents an advantage comparing with other tracer since it is emitted by occupants and it is inert (Hänninen, 2013). Since this method is focused on school classrooms that shifts strongly with time, as the opposed to what happens in ECCs, the method was adapted. The build-up curves used in this work corresponded to the periods that recorded changes on occupancy, e.g. lunch time.



Figure 4.2 – Geographical localization of the sampling site.

# 4.3.2 Samples collection

Two 2-weeks sampling campaigns were conducted – one during early fall (warm phase) and another throughout the winter (cold phase). The warm campaign occurred between  $5^{th}$  and  $15^{th}$  of October 2012 whereas the cold campaign was accomplished between  $28^{th}$  January and  $10^{th}$  of February 2013. Sampling was performed during 12 h periods during the day (8 AM – 8 PM) and during the night (8 PM – 8 AM). PM10 was collected in parallel in the ECC living-room and in the respective outdoor site by two TCR-Tecora® samplers operating at a flow rate of 2.3 m³ h¹¹. Particles were collected onto quartz filters with a diameter of 47 mm and a pore size of 2  $\mu$ m. During the sampling campaign, blank filters were treated the same way as regular samples. Figure 4.3 shows the location of the sampler in the outdoor, far away from any particular emission source. All measured species were homogeneously distributed; therefore, concentrations were corrected by subtracting the filter blank contents.



Figure 4.3 – Aerial view of the outdoor sampler.

# 4.3.3 Analysis of PM10

The collected filters were weighted using a Mettler Toledo balance with 0.1  $\mu g$  readability. The balance was placed in a controlled clean room (class 10,000) at a temperature of  $20\pm1$ °C and a relative humidity of  $50\pm5$ %. Before being weighted, filters were allowed to be equilibrated during 24 hours in the same room. When observed variations were less than 5  $\mu g$ , filters were weighted before and after sampling and mass was obtained as the average of three measurements.

Each filter was cut into three parts, controlled by weight, and each third was analysed by a different technique: a Thermal Optical technique for Organic Carbon (OC) and Elemental Carbon (EC) analysis; Ion Chromatography for the determination of inorganic water soluble ions and Instrumental Neutron Activation Analysis (INAA) for the elemental characterisation of particles.

A Thermal/Optical Carbon Aerosol Analyser (Sunset Laboratory) operating on the NIOSH Method 5040 was used to analyse OC and EC. The instrument's detection limit is 0.2  $\rm ugC/cm^2$  and the analytical uncertainty is equal to  $\pm$  (concentration\*0.05) + instrument blank concentration.

One third of the exposed quartz fiber filters were used for the determination of water-soluble inorganic ions. The filters were extracted with distilled deionised water by ultrasonic and mechanical shaking. The aqueous extract was analysed for anions (Cl<sup>-</sup>,  $SO_4^{2^-}$  and  $NO^{3^-}$ ) and cations (NH<sub>4</sub><sup>+</sup>, Na<sup>+</sup>, K<sup>+</sup>, Mg<sup>2+</sup>, Ca<sup>2+</sup>) using a DIONEX ICS-1100 chromatographic system, with IonPac AS22 (4mm) and IonPac CS12A (4mm) columns, respectively. Limit of detection (LOD) ranged from 0.01 to 0.11 µg.ml<sup>-1</sup> for anions and from 0.01 to 0.33 µg.ml<sup>-1</sup> for cations. Limit of quantification (LOQ) ranged from 0.03 to

 $0.36 \,\mu g.ml^{-1}$  for anions and from 0.01 to 1.08  $\mu g/ml$  for cations. Uncertainty (95%, k=2) ranged from 5.6 to 9.3% and from 2.6 to 10.7% for anions and cations, respectively.

One third of the filter was rolled up, put into aluminium foil and irradiated at the Portuguese Research Reactor (nominal power: 1MW) during 6 h. After the irradiation, filters were removed from the aluminium foil and were inserted in polyethylene containers. Samples were measured during 7-12 hours after 2-5 days and 4 weeks of decay, in a high resolution coaxial germanium detector, associated to an ORTEC® Automatic Sample Changer. A comparator – Al-0.1% Au alloy disk with a thickness of 125  $\mu$ m and a diameter of 0.5 cm – was co-irradiated with the samples for the application of the  $k_0$ -INAA methodology (Freitas et al., 2003, Freitas et al, 2004, Almeida et al., 2013a). Gamma Vision for Windows Model A66-B32, Version 6.08 was used to evaluate the net peak areas, the elemental concentrations were determined through  $k_0$ -standardized Instrumental Neutron Activation Analysis ( $k_0$ -INAA), and calculations were performed with the  $k_0$ -IAEA software (version 5.22). The agreement of the results was assessed by calculating the rations between the average results and the certified values from NIST-1633a, Coal Fly Ash, and revealed an agreement of  $\pm 12\%$  (Dung et al, 2010, Almeida et al, 2014b).

# 4.3.4 Meteorological data

Meteorological data from a weather station near the sampling site was a courtesy of Portuguese Institute of the Ocean and the Atmosphere. Data of temperature, relative humidity, wind direction, velocity wind and total amount of precipitation were kindly provided.

The wind roses and the pollutant dispersion maps were performed by Openair project software (NERC).

# 4.3.5 Statistical analysis

Principal Component Analysis (PCA) is a method that can be used for source apportionment. Sources categories for PM10 components were identified by means of PCA using Statistica® software. This was performed by using the orthogonal transformation method with Varimax rotation and retention of principal components whose eigenvalues were greater than unity. Factor loadings indicate the correlation of each pollutant species with each component and are related to the source emission composition. Only the species quantified in more than 80% of the samples were retained for PCA analysis. For data below detection limit, half of the detection limit values were used for calculations. PCA was performed with and without samples with outliers. The conclusions obtained were identical, therefore all samples were considered.

The crustal enrichment factor method was used to evaluate the strength of the crustal and non-crustal origin of the elements in aerosols. Enrichment factors, using Sc as a crustal

reference element (EF<sub>Sc</sub>) were calculated based on Mason and Moore (1982) and Taylor (1964) soil composition and on Equation 4.1:

$$EF_{Sc} = \frac{\frac{\left(\frac{X}{Sc}\right)_{aerosol}}{\left(\frac{X}{Sc}\right)_{crust}} \tag{4.1}$$

where,  $[X]_{aerosol}$  is the concentration of the element X in aerosol;  $[Sc]_{aerosol}$  is the concentration of Sc in aerosol, which was the element assumed to be uniquely characteristic of the soil;  $[X]_{crust}$  is the concentration of the X element in soil;  $[Sc]_{crust}$  is the Sc concentration in soil. Given the local variation in soil composition,  $EF_{Sc} > 10$  suggests that a significant fraction of the element is contributed by non-crustal sources (Chao et al. 2002).

Statistical calculations were performed by Statistica<sup>®</sup> software. The analysis of variance of results were performed by non-parametric statistics for a significance level of 0.05, the Wilcoxon Matched pairs for dependent groups (differences between pairs indoor and outdoor) and Mann-Whitney U Test for binary independent groups (differences between day and night and warm and cold campaigns). The Spearman test was also used to calculate the correlations between indoor and outdoor PM10 components concentration. It yields a statement of the degree of interdependence of the scores of the indoor and outdoor values.

## 4.4 Results and discussion

# 4.4.1 Meteorological conditions

Table 4.1 presents the basic statistics of the meteorological conditions during both campaigns. The variables temperature, humidity and wind speed presented statistical differences between both campaigns (p-values = 0.000, 0.040 and 0.000, respectively). The greatest difference was observed for the temperature, with a variance of 8.2°C between campaigns. On average, there were no significant differences between campaigns regarding the rainfall (p-value >0.05).

Table 4.1 – Basic statistics for the daily values of temperature (Temp), relative humidity (RH), wind speed (Ws) and rainfall (Rf) during the warm and cold campaign.

		Warm Campaign	Cold Campaign
	Ave	19	11
Temp	Std	3.1	2.5
(°C)	Min	13	6.0
	Max	28	18
	Ave	72	72
RH	Std	14	14
(%)	Min	31	14
	Max	98	100
	Ave	2.4	3.1
$\mathbf{W}\mathbf{s}$	Std	1.4	1.8
$(m.s^{-1})$	Min	0.0	0.0
	Max	7.7	10
	Ave	0.001	0.001
Rf	Std	0.02	0.02
(mm)	Min	0.0	0.0
	Max	0.4	0.5

This information was used as one inputs used to build the dispersion maps.

## 4.4.2 PM10 mass concentration

PM10 concentration was measured in the selected micro-environments in order to evaluate the ECC contamination and to identify the main emission sources. Figure 4.4 summarizes the results measured in both seasons and environments. In warm campaign, outdoor and indoor PM10 levels presented an average of 26  $\mu$ g.m<sup>-3</sup> in both environments. In cold campaign PM10 average concentration was 29  $\mu$ g.m<sup>-3</sup> and 28  $\mu$ g.m<sup>-3</sup> for the indoor and outdoor environment, respectively. Statistical analysis showed that there were not statistical differences between seasons and environments. In average, PM10 concentrations measured during the day were significantly higher than those during the night (p-value = 0.001), in both campaigns.

In average, indoor PM10 concentrations did not exceed the recommended value of 50 µg.m<sup>-3</sup> defined by the Portuguese legislation for indoor air quality (Portaria 353-A/2013). The results obtained in this work were at same range of values determined in London houses where indoor PM10 levels during spring and winter were 24 µg.m<sup>-3</sup> and 29 µg.m<sup>-3</sup>, respectively (Wheeler, et al., 2000). Also, similar results were obtained in a previous study conducted in ECCs' living-rooms during autumn and winter showed comparative concentrations (Almeida-Silva et al., 2014a). A study developed in UK residences

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presented similar results with PM10 average concentrations measured in indoor environment (Nasir et al., 2013). However, PM10 concentration levels measured inside this ECC were lower compared to the majority of the studies conducted in different public indoor micro-environments. Indeed, in this indoor environment the movement of people is limited since most of the institutionalized elders were semi-autonomous. Consequently, the possibility of re-suspension of dust was lower comparing with other crowded indoor environments, such as hospitals, schools, fitness centers and offices (Slexakova, et al, 2012; Canha, et al., 2012a; Ramos et al, 2014, 2015). Besides the low concentrations measured in this study, it is well-known that PM10 enhances adverse health effects and it is unclear whether a threshold concentration exists for PM below which no effects on health are likely. Moreover, elderly population not only is more susceptible to air pollutants but also spend most of their time in these micro-environments.

Outdoor PM10 average values did not exceeded the daily PM10 limit value of 50  $\mu g.m^{-3}$  defined by the European Directive 2008/50/EC (EU, 2008), the U.S. Environmental Protection Agency health-based air quality PM10 annual standard of 150  $\mu g.m^{-3}$  (USEPA, 2008), nor the World Health Organization PM10 annual standard of 50  $\mu g.m^{-3}$  (WHO, 2005).

The relation between indoor and outdoor concentrations is an important indicator of the relative strength of outdoor sources in the indoor environment (Massey, et al., 2012), once indoor PM concentrations could be affected by 1) penetration from outdoors into the buildings; 2) the outdoor air brought in by natural ventilation; and 3) indoor sources (Estoková, et al., 2010; Madureira, et al., 2012).

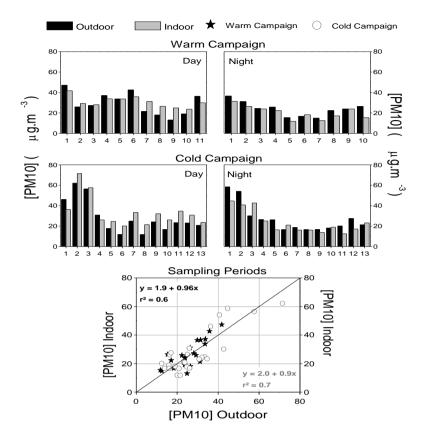


Figure 4.4 – PM10 concentrations for each sampling campaign and ratio indoor/outdoor for warm and cold campaigns.

Figure 4.4 shows that there was a significant contribution of the ambient atmosphere to the indoor levels due to the observed correlations between indoor and outdoor concentrations ( $r^2 = 0.6$  and 0.7 for warm and cold campaign, respectively). In average, I/O ratio for warm and cold campaigns was similar, presenting values of 1.1 and 1.0, respectively. These findings clearly indicated that there was a large contribution of the ambient atmosphere to the indoor levels. Several studies presented similar results, e.g. the study of Madureira (2012) which presented an I/O ratio around 1.1 for the period of the day in dwellings, as well as, Lawson study (2011). Higher I/O ratios were observed in micro-environments with high occupancy, as well as, when intense human activities took place inside the rooms, suggesting a greater contribution of indoor emission sources (Diapouli, et al., 2008).

# 4.4.2.1 Differences between warm and cold campaigns

As shown in Figure 4.5, during the warm campaign the winds come essentially from the 3<sup>rd</sup> and 4<sup>th</sup> quadrants, whereas the highest PM10 concentrations were observed in 2<sup>nd</sup> and 3<sup>rd</sup> quadrant and associated with highest wind speed. During the cold campaign, the highest PM10 concentrations occurred mainly with the lowest wind speeds while being associated with the same quadrants, 2<sup>nd</sup> and 3<sup>rd</sup>. These results showed that the highways and the International Airport of Lisbon, which are located in south west and south, respectively, had an important contribution to particles concentration in the studied area. Two different studies developed in schools located near roadways, showed that the traffic is the main source of particles (Braniš, et al., 2011; McConnell, et al., 2010). Also, previous works demonstrated that traffic is responsible by the production of secondary particles, principally associated with road vehicles exhaust, and by the emission of primary particles resulting from tires and brake wear and soil re-suspension (Almeida et al., 2009b). Figure 4.5 indicates a potential contribution of a small industrial area (e.g. car repairs, company of transports of merchandises and warehouses) that exists northeast of the sampling point.

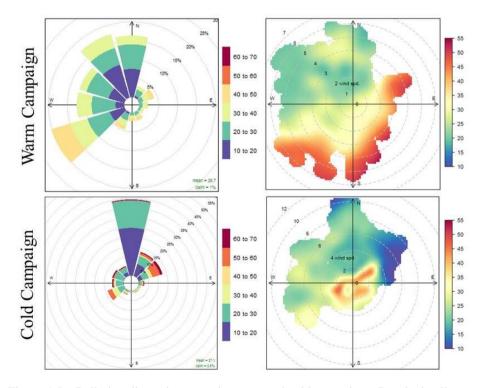


Figure 4.5 – Pollution dispersion maps in warm and cold campaigns. Results in all maps are presented in  $\mu g.m^{-3}$ .

## 4.4.2.2 Differences between day and night

The direction of the wind was similar during the day and night. In average, northern winds were most frequent, followed by eastern and southern winds, while the strongest winds occurred essentially during the day (in the order of 12 m.s<sup>-1</sup>). The greatest difference between day and night was that approximately 20% of the winds come from the 4<sup>th</sup> quadrant during the day.

According to Costa (2004) the movement of thermal depression in the Iberian Peninsula gives rise to the wind from the north or northwest across the west coast with increasing intensity throughout the day, reaching a maximum at the end of the afternoon. During the night the northern winds loose intensity.

In pollutant dispersion maps, PM10 concentrations are shown to vary with wind speed and wind direction. As can be derived from Figure 4.6, PM10 concentration was higher during the day, mainly in 2<sup>nd</sup> and 3<sup>rd</sup> quadrant. When the winds come from the south, they brought larger concentrations of PM10 which did not occur when they were originated from the north. Once again, results proved the influence of the airport and the highways to the high PM concentrations measured in the studied area.

Results also showed that PM10 concentrations were significantly higher during the day than night (*p*-value = 0.001), while presented a day/night ratio of 1.15. The highest concentrations noticed during the day may be attributed to the fact that the International Airport of Lisbon (the Portugal's largest airport, according to the annual report of statistical air traffic of ANA – Airports of Portugal) was characterized by more 95% of traffic during the day than night (ANA, 2011).

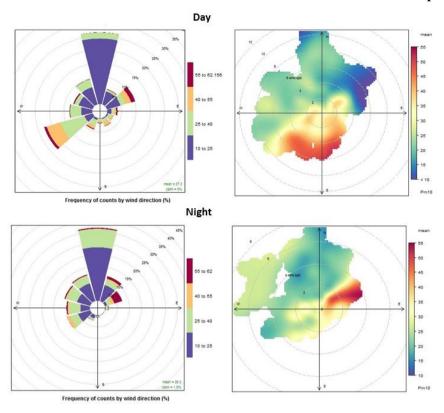


Figure 4.6 – Pollution dispersion maps comparing day and night. Results in all maps are presented in  $\mu g.m^{-3}$ .

During the night period, it is possible to observe high concentrations of PM10 when the winds were coming from NE. At the northeast of the ECC, there is an area with garages (car repairs), company of transports of merchandises and warehouses which may justify those high values, not only because they are open until late hours but also due to the high PM10 emissions.

# 4.4.3 PM composition

Table 4.2 summarizes the average and the standard deviation of PM10 components (carbonaceous, water soluble ions and trace elements).

On average, the sum of the indoor PM10 components measured in this work explained 57% and 53% of the total PM10 mass measured by gravimetry in warm and cold campaigns, respectively. The remainder percentage might be associated with organic compounds and other PM species that were not assessed in this work. The major component measured in

indoor PM10 was the carbonaceous fraction (OC and EC), representing on average 31% (STD=14) in warm campaign and 30% (STD=5) in cold campaign of the total indoor PM10 mass measured by gravimetry.

The results showed that OC concentration was significantly higher indoors (*p*-value = 0.0002), could be attributed to sub-micrometre fragments of paper, skin debris, clothing fibers, cleaning products and waxes produced indoors (Alves et al., 2014). On average, OC accounted for 24% (STD=9) of the mass of PM10 indoors, whereas a lower mass fraction was found outdoors [14% (STD=5)], which represented an average I/O ratio of 1.8. The EC concentration was not significantly higher indoors compared to the outdoor one (*p*-value = 0.910) while the I/O ratio for EC presented lower values, probably due to the influence of traffic.

Organic to elemental carbon (OC/EC) ratios exceeding 2.0 have been used as an indicator for the presence of secondary organic aerosols (SOA) (Chow et al., 1996) in urban areas where traffic emissions had a significant contribution to the EC levels. In this study, the average indoor OC/EC ratio was 6.0 and 7.6 in warm and cold campaign respectively. This shows a significant increase of OC levels that could be attributed to primary indoor sources and also to SOA production. The formation of SOA in indoor environments was demonstrated and confirmed in test chambers experiments, done by Aoki and Tanabe (2007). Sportion of semi-volatile organic compounds could be as well as important indoor source of organic matter. Thus, the combination of active indoor sources, sorptive processes and SOA formation leaded to an enrichment of the indoor particles in OC and to high OC/EC ratios inside the ECC (Alves et al., 2014). Furthermore, the cleaning routines and, consequently, the use of detergents everyday may contribute to the production of SOA. According to Ji and Zhao (2015) the SOA contribution to the indoor PM2.5 concentration might be very small, less than 3%. Considering these two parameters, significant differences between concentrations measured day and night were not observed (p-value > 0.05).

Table 4.2 – PM10 concentration and PM10 composition measured in both campaigns.

•		Warm Ca	mpaign		Cold Campaign				
	Indoor		Outdoor		Indoor		Outdoor		
	Day	Night	Day	Night	Day	Night	Day	Night	
PM10 [μg.m <sup>-3</sup> (STD)]	31(5)	20(6)	29(10)	23(7)	35(14)	24(11)	29(16)	27(14)	
Carbonaceous com	ponents [µg.m	-3(STD)]							
$\mathbf{OC}^{*w,c}$	6.3(1.1)	4.8(1.7)	4.4(1.5)	3.3(1.8)	7.6(1.8)	6.3(2.3)	3.9(3.1)	3.6(3.2)	
EC	1.8(1.4)	1.2(0.9)	2.2(1.1)	1.5(1.1)	1.5(1.1)	2.0(2.5)	1.8(1.5)	1.7(1.8)	
TC	8.1(2.3)	6.1(2.3)	6.6(2.3)	4.8(2.8)	9.1(2.5)	8.3(4.5)	6.0(4.2)	5.3(4.8)	
Cations [µg.m <sup>-3</sup> (STD)]									
Na <sup>+</sup> *w,c	0.95(0.75)	1.2(0.8)	1.04(0.94)	1.1(0.6)	0.94(0.49)	0.81(0.53)	1.5(1.2)	2.2(1.4)	
NH <sub>4</sub> <sup>+</sup>	0.34(0.23)	0.42(0.29)	0.35(0.22)	0.7(0.5)	1.3(1.3)	0.78(0.82)	1.0(1.3)	0.6(0.6)	
$\mathbf{K}^{+} \Psi$	0.09(0.05)	0.13(0.2)	0.16(0.14)	0.11(0.1)	0.29(0.23)	0.16(0.14)	0.25(0.24)	0.16(0.13)	
Mg <sup>2+ *w,c</sup>	0.13(0.08)	0.15(0.08)	0.18(0.14)	0.2(0.06)	0.13(0.06)	0.13(0.08)	0.21(0.14)	0.28(0.17)	
Ca <sup>2+ δ, *w,c</sup>	0.77(0.37)	0.51(0.28)	0.99(0.37)	0.74(0.29)	0.76(0.38)	0.4(0.3)	0.88(0.51)	0.53(0.45)	
Anions [µg.m <sup>-3</sup> (STD)]									
Cl <sup>-</sup> ψ, *w,c	1.3(1.1)	0.96(0.78)	1.8(1.9)	2.7(3.5)	1.3(0.8)	1.1(0.85)	3.4(3.2)	4.4(3.3)	
NO <sup>3-*w,c</sup>	1.5(0.8)	1.4(0.9)	1.4(0.6)	2.9(2.0)	1.9(2.0)	1.4(1.6)	3.2(3.8)	2.7(2.0)	
SO <sub>4</sub> <sup>2</sup> -	1.9(1.6)	2.1(1.5)	1.6(1.0)	2.1(1.1)	2.2(3.1)	1.5(1.7)	2.3(2.7)	2.3(1.9)	
Elements [ng.m <sup>-3</sup> (STD	<b>)</b> )]								
As*c	0.51(0.21)	0.41(0.19)	0.7(0.5)	0.33(0.19)	0.84(0.76)	0.8(0.7)	0.6(0.4)	0.55(0.23)	
Ce	0.7(0.3)	0.6(0.4)	1.0(0.6)	0.6(0.4)	0.6(0.4)	0.6(0.2)	0.7(0.4)	0.47(0.34)	
Co*c	0.1(0.07)	0.07(0.06)	0.35(0.23)	0.26(0.12)	0.2(0.1)	0.3(0.2)	0.4(0.4)	0.12(0.05)	
Cr*w,c	1.9(1.6)	2.4(4.2)	6.1(4.4)	3.7(2.4)	3.0(1.8)	1.4(0.2)	1.8(1.9)	1.3(1.1)	
Fe*w,c o	310(210)	200(130)	600(380)	330(180)	170(130)	200(210)	460(290)	240(180)	
K	191(60)	120(50)	230(130)	140(80)	280(200)	200(180)	100(120)	160(140)	
Na <sup>*w,c</sup>	940(660)	640(280)	1044(681)	1200(1080)	620(380)	480(300)	990(630)	1100(970)	
Sb <sup>δ</sup>	2.2(1.9)	1.0(0.9)	3.0(2.4)	1.7(0.9)	1.6(1.4)	1.9(3.2)	4.2(7.4)	1.2(1.6)	
Sc*w,c	0.04(0.01)	0.03(0.02)	0.1(0.04)	0.07(0.02)	0.07(0.01)	0.06(0.02)	0.06(0.02)	0.05(0.02)	
Sm	0.06(0.03)	0.05(0.03)	0.07(0.04)	0.05(0.04)	0.16(0.27)	0.08(0.07)	0.04(0.04)	0.1(0.06)	
Zn <sup>*c δ ψ</sup>	35(15)	12(8)	40(22)	15(6)	28(16)	1.9(3.2)	16(13)	10 (11)	

<sup>\*</sup>w Significant difference between indoor and outdoor in warm campaign: for p-value < 0.05; \*c Significant difference between indoor and outdoor in cold campaign: for p-value < 0.05;  $\delta$  Significant difference between day and night: for p-value < 0.05;  $\psi$  Significant difference between warm and cold campaigns: for p-value < 0.05.

Ions comprised the major component of outdoor PM10, representing on average 35% (STD=19) and 45% (STD=24) of the total indoor PM10 mass measured by gravimetry in warm and cold campaign, respectively. Results showed that  $Cl^-$  and  $K^+$  presented significantly higher concentrations in the cold campaign (p-values = 0.019 and 0.011, respectively).

On average, ions accounted for 22% (STD=10) of the mass of PM10 indoors, whereas a higher mass fraction was found outdoors (40% (STD= 22), which represented an average I/O ratio of 0.6. Cl<sup>-</sup>, NO<sub>3</sub><sup>-</sup>, Na<sup>+</sup>, Ca<sup>2-</sup>, Mg<sup>2+</sup> concentrations were significantly higher outdoor compared to the indoor one (p-value < 0.05), presenting an I/O ratio of 0.35, 0.60, 0.60, 0.59 and 0.80, respectively. These ions are commonly associated with sea spray, secondary aerosols and also combustions (Almeida et al., 2005).

The element fraction for the total PM10 was 3.7% and 6.4% for indoor and outdoor, respectively. Fe, K and Na were the most abundant elements both indoors and outdoors. Zinc presented significantly higher concentrations in warm campaign compared to the cold campaign (p-value = 0.017). Results showed that As, Cr and Zn presented significantly higher concentrations in indoor than outdoor air (p-value < 0.05), indicating the existence of indoor sources for these elements. Nevertheless, according to Figure 4.7 the correlation between indoor/outdoor for these elements were only significant for As and Zn ( $\rho$  = 0.62 and p-value < 0.05 for both).



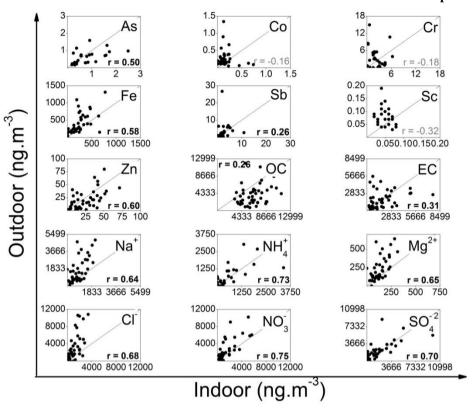


Figure 4.7 – Relation of PM10 components' concentration in indoor environment and corresponding outdoor environment. The correlation coefficient (r) between the mean of indoor values and its outdoor are presented in the figure (bold values are statistically significant: p-values < 0.05).

Enrichment factors for these elements (and also for Sb) indicated that they were enriched in relation to the soil reference. Outdoor As, Cr, Sb and Zn levels are typically associated with traffic, mainly with re-suspension/dispersion of road dust, tire wear, and break wear (Almeida-Silva et al., 2011). Besides the great contribution of outdoor pollutants to indoor environment, several studies had already observed significantly higher concentrations of Zn emitted by indoor products applied e.g. to protect steel, walls, wood surfaces, doors and windows (Avigo et al., 2008).

An opposite behaviour was observed for Fe, Na and Sc which present significantly higher concentrations outdoors than indoors (p-values < 0.05). Figure 4.8 also shows that those elements were not enriched indicating a provenience of these elements from natural sources: Fe and Sc from the soil; and Na from sea-salt spray.

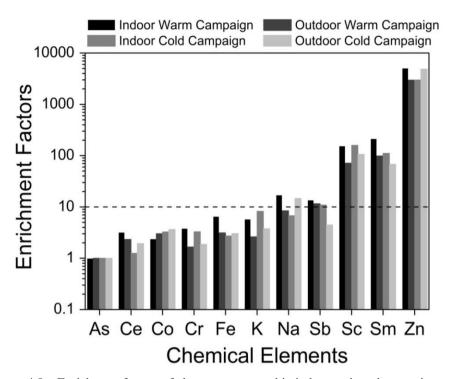


Figure 4.8 – Enrichment factors of elements measured in indoor and outdoor environments in both campaigns.

Fe, Sb and Zn were the three elements that presented significantly higher concentrations during the day (*p*-value = 0.000004; 0.012; 0.037, respectively). The elements that presented higher concentration during the day are essentially associated with dust resuspension (Fe) (Almeida-Silva et al., 2011; Almeida et al, 2008) and dispersed road dust, exhaust break wear, tires and motor oil (Zn and Sb) (Sternbeck et al., 2002; Zechmeister et al., 2005, 2006; Ayrault et al., 2010, Almeida et al., 2009b). These results prove the influence of the airport and the highways to the high concentration of outdoor anthropogenic sources measured in the studied area; because, not only the number of vehicles decrease during the night period but also the air traffic (wind direction and velocity were explained in sub-chapter 4.4.2.2).

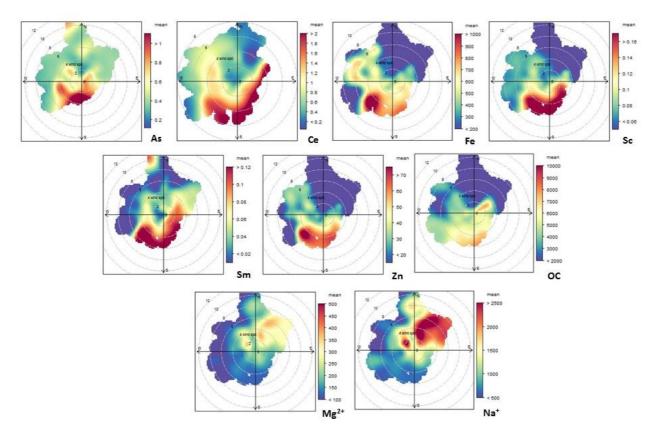


Figure 4.9 – Pollution dispersion maps for the selected compounds: As, Ce, Fe, K, Sc, Sm, Zn, OC,  $Mg^{2+}$  and  $Na^{+}$ . Results in all maps are presented in  $\mu g.m^{-3}$ .

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Figure 4.9 was built in order to understand the provenience and the spatial distribution of the PM10 compounds (the discussion will be done with a selection of PM10 compounds).

The results showed that the highways and the Lisbon airport, which are located in south west and south, respectively, have an important contribution for the concentration of PM10 compounds in the studied area. All of the represented compounds presented a day/night ratio higher than 1 (Table 4.3), which might support its association with highways and the International Airport of Lisbon (since its emissions decrease during the night).

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Table 4.3 – Indoor/outdoor (I/O) and day and night (D/N) ratio's for each PM10 component. The cells fulfil with grey presented ratios higher than 1. The values in bold presented significant differences between indoor/outdoor and/or day/night. The \* represent the components that presented significant differences between campaigns (warm and cold). In all cases the significance value is for a p-values < 0.05.

	Warm Campaign	Cold Campaign Warm Campaign			Cold Campaign		
	I/O	I/O	Indoor D/N	Outdoor D/N	Indoor D/N	Outdoor D/N	
ос	1.45	1.75	1.33	1.33	1.21	1.18	
EC	0.80	1.03	1.45	1.50	0.73	1.05	
Na <sup>+</sup>	1.05	0.42	0.80	0.68	1.15	0.70	
NH <sub>4</sub> <sup>+</sup>	0.67	0.91	0.82	0.55	1.72	1.63	
K**	0.81	0.88	0.74	1.44	1.88	1.56	
Mg <sup>2+</sup>	0.83	0.48	0.89	0.74	0.97	0.76	
Ca <sup>2+</sup>	0.76	0.86	1.51	1.38	1.58	1.67	
Cl-*	0.85	0.27	1.26	0.67	1.15	0.76	
NO <sup>3-</sup>	0.72	0.63	0.97	0.50	1.23	1.20	
SO <sub>4</sub> <sup>2</sup> -	1.08	0.76	0.74	0.75	1.51	0.96	
As	0.80	1.58	1.27	2.11	1.02	1.11	
Ce	0.77	0.95	1.06	1.75	1.05	1.55	
Со	0.29	0.95	1.46	1.35	0.66	2.77	
Cr	0.43	2.59	0.82	1.64	0.92	1.34	
Fe	0.51	0.68	1.54	1.83	0.82	1.94	
K	0.85	1.87	1.59	1.67	1.35	0.63	
Na	0.75	0.49	1.48	0.87	1.31	0.87	
Sb	0.63	0.65	2.12	1.79	0.84	3.42	
Sc	0.37	1.06	1.17	1.47	1.05	1.10	
Sm	0.81	2.31	1.15	1.39	1.96	0.44	
Zn*	0.81	1.73	2.91	2.74	1.67	1.54	

## 4.4.4 Source apportionment

PCA with varimax rotation method was applied for the identification of the possible pollutant sources of PM10 components. Five principal components (PC) were extracted and enough to explain 80% of the total variance. Table 4.4 presents the PCA results of PM10 components in indoor and outdoor. Factor loadings > 0.7 or < -0.7 (in bold and filled gray) are considered to be important, representing the contribution of each variance to the corresponding principal component.

In indoor sampling site, the first PC was associated with secondary aerosols, being characterized by NO<sub>3</sub><sup>-</sup>, SO<sub>4</sub><sup>2-</sup> and NH<sub>4</sub><sup>+</sup> (Almeida et al., 2005). The second PC was dominated by Sb, Zn and EC, explaining 21% of the variance. These elements were reported elsewhere to be defined as traffic fingerprints (Almeida-Silva et al., 2011; Calvo et al., 2013). Sea spray is clearly represented in PC3, by Cl<sup>-</sup> (0.90), Na<sup>+</sup> (0.91) and Mg<sup>2+</sup> (0.93). Co and Sc are two well-known crust elements (Calvo et al., 2013) and both are well correlated in PC4 (0.83 and 0.89, respectively). PC5 is defined as "Other" due to the only PM10 component that are represented is the Cr. This PC might be associated with an indoor source, since the I/O ratio for Cr was higher than 1. Moreover, the correlation between Cr indoor/outdoor was quite low and non-statistically significant (Figure 4.7).

In outdoor sampling site, the PC1 was also associated with secondary aerosols, presenting a very similar results comparing to PC1 in indoor sampling site. PC2 was dominated by components associated with sea spray: Cl<sup>-</sup> (0.96), Na<sup>+</sup> (0.98) and Mg<sup>2+</sup> (0.96); explaining 18% of the variance. In outdoor environment the traffic source were segmented in two: 1) re-suspension and 2) combustion. Each one explained 18% and 13% of the variance, respectively. This separation was due to the high correlation of Cr (0.79), Fe (0.79) and Zn (0.82) in PC3 and OC (0.86) and EC (0.81) in PC4. Finally, the PC5 was dominated by the contribution of Co, an element known to be related with the soil (Calvo et al., 2013).

Table 4.4 – Factor profile contributions to PM10 components occurrence obtained by PCA.

;	Indoor Outdoo									
	PC1	PC2	PC3	PC4	PC5	PC1	PC2	PC3	PC4	PC5
	Secondary	Traffic	Sea	Soil	Other	Secondary	Sea	Traffic (re-suspension)	OC/EC (combustion)	Soil
As	0.63	0.32	-0.16	0.38	-0.03	0.39	0.02	0.49	0.07	0.31
Co	0.13	0.08	0.08	0.83	0.21	0.05	-0.07	0.21	-0.12	0.86
Cr	0.27	0.15	0.24	0.14	0.71	-0.12	-0.2	0.79	0.06	-0.01
Fe	0.09	0.67	-0.16	0.08	0.58	0.2	-0.23	0.79	0.25	0.27
Sb	0.26	0.78	-0.16	0.03	0.2	0.56	0.1	0.58	-0.02	0.06
Sc	0.08	0.07	-0.14	0.89	-0.08	-0.1	-0.13	0.46	0.35	0.66
Zn	0.27	0.83	0.07	0.1	0.06	0.06	-0.05	0.82	0.28	0.31
ос	0.39	0.69	-0.01	0.3	-0.2	0.2	0.11	0.2	0.86	0.02
EC	0.18	0.83	-0.09	0.01	0.25	0.18	0	0.3	0.81	-0.13
Cl	-0.13	0.12	0.90	0.1	-0.19	-0.05	0.96	-0.13	0.01	-0.06
NO <sub>3</sub>	0.89	0.27	0.06	0.04	0.22	0.88	-0.15	0.07	0.16	-0.04
SO <sub>4</sub> <sup>2-</sup>	0.85	0.1	0.03	-0.08	0.39	0.89	0	0.08	0.22	0.03
K⁺	0.66	0.36	0.07	0.3	0.01	0.6	0.09	-0.27	0.46	0.25
Na⁺	0.04	-0.21	0.91	-0.12	0.27	-0.07	0.98	-0.1	0	-0.06
Ca <sup>2+</sup>	0.35	0.45	0.14	-0.03	0.58	0.41	-0.11	0.03	0.51	0.23
Mg <sup>2+</sup>	-0.01	-0.11	0.93	-0.08	0.19	-0.14	0.96	-0.09	0.04	-0.04
NH₄⁺	0.85	0.25	-0.2	0.08	0.12	0.85	-0.27	0.11	0.14	-0.07
Expl. Var. %	21%	21%	16%	11%	10%	20%	18%	18%	13%	9%

Comparing the factor profile contributions indoor with outdoor and considering the Figure 4.7 it is possible to observe that  $NO_3^-$ ,  $SO_4^{2-}$  and  $NH_4^+$  (associated with secondary aerosols) and Cl<sup>-</sup>, Na<sup>+</sup> and Mg<sup>2+</sup> (related to sea spray) demonstrated significant correlations between indoor and outdoor concentration, presenting p-values < 0.05. This finding indicates the existence of common indoor and outdoor sources for these elements, which supports the contribution of outdoor to indoor pollutant concentration. Cobalt and scandium, two elements related with the soil' source and both well correlated in PC4 and PC5 indoor and outdoor, respectively, did not presented significant correlations between indoor/outdoor (with p-values of 0.53 and 0.061, respectively). Other difference is the fact that traffic source were segmented in two PC: 1) re-suspension and 2) combustion; as it was referred previously. This segmentation in outdoor environment could be related with the existence of important outdoor sources nearby, such as the highway and the airport. Moreover, may contribute to the appearance of a source related to the re-suspension of dust (PC3 in outdoor) and another related to the fuel combustion (PC4 in outdoor), while in indoor environment all of these components were associated at the same PC (PC2 in indoor). Other great difference is the existence of a type of source defined as "Other" in indoor environment, related with Cr. The explanation was already documented.

This approach justify the need of a carefully choice of geographical location of ECC, in order not only to reduce indoor air pollutants that are originated from outdoor air by infiltration or natural ventilation but also to mitigate the elderly exposure to air pollutants.

### 4.5 Conclusions

Considering the results obtained in this work it was possible to conclude that:

- Outdoor has a significant contribution to indoor particulate matter concentrations;
- Outdoor PM10, Ca<sup>2+</sup>, Fe, Sb and Zn concentrations were significantly higher during the day than night;
- Both indoor and outdoor PM10 average concentration did not exceed the daily PM10 limit values;
- No significant differences were observed between seasons and environments;
- The greatest difference between day and night is justified by the fact that almost 20% of the winds come from the 4<sup>th</sup> quadrant during the day;
- PM10 concentration was significantly higher during the day, mainly in 2<sup>nd</sup> and 3<sup>rd</sup> quadrant;
- Carbonaceous fraction (OC and EC) represented 31% and 30% of indoor PM10 in cold and warm campaign, respectively;
- OC concentration was significantly higher indoors, being associated with submicrometer fragments of paper, skin debris, clothing fibers, cleaning products and waxes may;

#### **Chapter IV**

- Ions comprised the major components of outdoor PM10, representing on average 35% (±19) and 45% (±24) of the total indoor PM10 mass measured by gravimetry in warm and cold campaign, respectively;
- Cl and K presented significant higher concentrations in the cold campaign than warm campaign;
- The element fraction for the total PM10 was 3.7% and 6.4% for indoor and outdoor, respectively;
- Fe, K and Na were the most abundant elements both indoors and outdoors;
- Five PC's were extracted and enough to explain 80% of the total variance;
- Secondary aerosols, sea spray and soil were identified as common sources in both indoor and outdoor environment;
- Traffic was also identified as a source, but was separated in two PC's in outdoor environment (re-suspension and combustion);
- The highways and the airport (located less than 500 m of the ECC) have a great importance for both indoor and outdoor air quality.

In conclusion, the geographical localization of Elderly Care Centers is a very important issue. The existence of surrounding emission sources could lead to an increase of elderly exposure to air pollutants. Moreover, a carefully choice of materials, products and routines should be taken into account, in order to mitigate the exposure of this susceptible population to chemical pollutants. It is suggested that elderly supporters and ECC proprietors have training related to elderly health conditions, air pollution, chemical and physical pollutants, emission sources of air pollutants, etc. This approach plus their knowledge regarding the life and physical and psychological status of the elderly population will potentiate the elderly well-being and, probably, life expectancy.

### CHAPTER V. CONCLUSIONS

The current thesis improved the state of the art regarding elderly and ECCs, filling the gap of knowledge about elderly exposure to air pollutants, indoor air quality in ECCs and emission sources that may influence the elders, in Portugal. The general discussion and the final remarks will present the highlights provided from this thesis.

#### 5.1 General discussion & Final remarks

The basis of the increased sensitivity in elderly is not known but it is likely that it is linked to age-related impaired function of the lung and deteriorated health status. Old people will continuous to experience more chronic conditions, more activity limitations and disability related to chronic disease, having more chronic conditions than younger persons. This phenomenon is influenced by a number of factors: environmental factors, genetic predisposition, disease agents, and lifestyle choices. So, it is crucial to act upstream older ages, creating and applying the best preventive healthcare actions available, in order to mitigate as much as possible individual factors that could promote adverse health effects.

The study design did not allow to assess causality. Nevertheless, it was crucial to better understand the elderly life occupancy pattern – 95% of their time was spent indoors, splitted between bedroom and living-room – and to prove that exposure to indoor air pollutants is a current problem that is necessary to be aware. Additionally, the thesis showed that natural ventilation did not achieve the minimum air renovation to provide good IAQ. This fact lead to high concentrations of CO<sub>2</sub>, which exceeded the CO<sub>2</sub> limit values provided from Portuguese legislation. Other problematic air pollutant was the VOC. Essentially emitted by (e.g.) furniture, cabinetry, vinyl wall coverings, adhesives, and cleaning products, these pollutant presented high concentrations in 75% of the analysed indoor micro-environments. Fungal concentrations were also assessed and it was consider the necessity to perform further and long studies to better understand the elderly exposure to this bio-aerosols.

Particulate matter was the pollutant studied in a deeper and detailed way in this thesis. Temporal concentrations allowed the identification of hot-spots or problematic periods of the day. In bedroom, the worst period of the day was associated with the elders uprising. In living-room, the highest PM concentration was associated with the people movement at specific periods, such as breakfast and lunch time. Chemically characterization permitted identifies indoor and outdoor emission sources, promoting the creation of mitigation actions to reduce its emissions. This thesis also showed the different micro-environments' contributions to elderly exposure and dose. Although, the highest PM concentrations were registered inside living-rooms, elders spent the majority of their time in bedrooms. Moreover, this thesis revealed the importance of a coherent and strategic choice of geographical localization of Elderly Care Centers, due to the pollutants emitted by possible

surrounding emission sources that penetrate in indoor environments and contribute for the increasing of elders' exposure to those air pollutants.

Even at low concentrations, indoor air pollutants present in ECCs may have important impacts on the health of elderly living there permanently, due to their long exposure periods. In fact, from the pathophysiological point of view, being exposed for 1 h every day to a certain pollutant for 20 days is different from being exposed for 20 h to the same pollutant for only 1 day.

#### **5.2 Future research**

Sparse population data had showed that elderly are particularly susceptible to air pollutants, since it has been suggested that air pollution could precipitate health status in elderly population due to their frailty. It seems urgent to identify those situations to promote the best preventive actions, in order to potentiate and improve the elderly wellbeing and lifestyle. To achieve this goal I believe that is necessary to:

- create a transdisciplinary collaborations to expand the number of sample and the number of measured agents;
- assess individual exposure and health outcomes;
- better understand the biological interactions between air pollutants and the biological pathways;
- develop statistical models adapted to concentrations of a high number of correlated pollutants allowing an adequate selection among the measured pollutants.

Moreover, several works had affirmed the necessity of future studies considering the size-fractionated PM, in order to improve identification of determinants of PM exposure linked with time-activity patterns and intermittent sources. This approach can be promoted by real-time instruments and personal exposure monitoring (Urso et al., 2015). However, this exposure needs to be assessed using objective measurements, taking into account the specification of each emission source and its influence. Additionally, suitable, noiseless, cheap and weightless personal samplers should be used in order to get the most realistic personal exposure to air pollutants and, at the same time, mitigate the annoyance of the subject.

Due to the complexity of indoor air pollution and its variability with time, estimation of risk associated with exposure to complex mixture and the generalization of the obtained results is rarely done or feasible. Therefore, to calculate risk assessment for chemical mixtures or for combine effects, due to concomitant exposure to different chemicals through different routes, seems urgent.

Finally, further studies are needed to investigate not only the environmental impact of risk factors in elderly population and their homes (in this particular case: Elderly Care Centers), but also to underline biological, toxicological and physiological mechanisms.

The thesis highlighted the importance of study the elderly population since their greater susceptibility to air pollutants, However, as it was described in Chapter I (1.1 Elderly population – an emerging concern) the concerns related to elderly people are not only restricted to environmental and health problems. In fact, there is a huge societal issue regarding this subject. The worldwide population is getting older (> 60 years), had increased 500 million in twelve years and estimated to be 18% of the total population in 2050. This, amongst low bird rates promote an inverse in age pyramid, and, consequently, may lead to fewer men and women of working ages, and hence a smaller tax base to finance social security payments. As longevity increases, the proportion of people who are old will grow the fastest and their demand for care and support will increase, particularly due to age-related diseases. In addition, demographic changes mean that for some there is a lack of family support, which will place further burden on the long term care market in the future. According to the American Council of Life Insurance (1998), almost 91 billion dollars were spent for long-term care for the elderly in 1995. So, it seems important to create and develop new and integrated projects that merge, not only the environmental concerns but also the societal issues, in order to promote a better life to old people, a better (and cheaper) care and nursing system and to potentiate the relations between different populations – from children to elderly.PM

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#### **SUMMARY**

This Thesis focuses on the estimation of the human exposure to air pollutants, and gives special attention to one of the most susceptible groups in the general population - elders. To fulfil the goal the work was conducted following the risk assessment paradigm and, consequently, was divided into 5 tasks: 1) characterization of the indoor air quality in (selected) Elderly Care Centers, thereby evaluating an mixture of indoor physical, chemical and biological pollutants; 2) assessment of the daily integrated exposure and inhaled dose; 3) determination of the personal daily exposure; 4) modelled estimation of the particulate matter (PM) deposited dose in the respiratory tract; 5) source apportionment of indoor PM in an Elderly Care Center.

For this thesis was important to better understand the elderly life occupancy pattern: 95% of their time was spent indoors, splitted between bedroom and living-room.

Chapter 2 describes the characteristics of ten selected Elderly Care Centers and its 384 institutionalized elders, located in Lisbon and Loures, Portugal. Some problematic pollutants were identified, such as carbon dioxide, volatile organic compound and particles. Taking PM into account the highest impact period of the day was associated with the elders getting out of bed in the morning and with the people movement at specific periods, such as breakfast and lunch time, in bedrooms and living-rooms, respectively: temporal concentration patterns allowed the identification of hot-spots or problematic periods of the day. Chemical characterization permitted the identification of indoor and outdoor emission sources, and permitted the set-up of mitigation actions to reduce their emissions. The results showed that, although elders may live in the same area, their exposure and inhaled doses can differ significantly. Moreover, the data also showed that an accurate measurement of integrated exposure is essential to provide an adequate evaluation of the particles dose-response relation. The outcomes were important i) to understand the critical areas inside the residences, ii) to identify the microenvironments with highest impact on elderly exposure and dose and iii) also to recognise the importance of the geographical localization of Elderly Care Centers.

In Chapter 3 PM exposure and dose were calculated for autonomous and institutionalized elders. Autonomous elders carried on a real-time PM monitor during four 24 h measuring campaigns and fulfilled an activity diary, simultaneously. Subsequently, the PM deposited dose was assessed using a computational model that estimates transport and deposition of particles into the human respiratory tract. In general, the deposition fraction was higher in males than in females for the same activity level and increased with activity level. The results were important to understand the relevant areas inside the residences where elders are most exposed, to suggest effective mitigation strategies, and thus to reduce PM exposure and adverse health effects to this very sensitive age group.

Chapter 4 presents the results of the indoor PM source apportionment study 'in a selected Elderly Care Center. To this end, two 2-weeks sampling campaigns were conducted to

collect PM10. The elemental composition, ions, and the organic and elemental carbon were determined in order to identify emission sources. Outdoor  $PM_{10}$  concentrations were significantly higher during day-time than at night (p-value <0.05), as well as  $Ca^{-2}$ , Fe, Sb and Zn. Both indoor and outdoor  $PM_{10}$  averaged concentrations did not exceed existing guidelines and there were no significantly differences between seasons. The contribution of indoor and outdoor sources was assessed by Principal Component Analysis and showed the importance of the highways and the airport located less than 500 m of the Elderly Care Center.

#### **SAMENVATTING**

Dit proefschrift richt zich op de bepaling van humane blootstelling van luchtverontreiniging, met speciale aandacht voor een van de meest gevoelige groepen in de bevolking – ouderen. Om aan dit doel te kunnen beantwoorden werd de studie uitgevoerd in lijn met het risico-bepalings-model, en onderverdeeld in 5 taken: 1) karakterisering van de kwaliteit van lucht binnenshuis in geselecteerde verzorgingstehuizen, waarbij de mix aan fysische, chemische en biologische verontreinigingen werd ge-evalueerd, 2) bepaling van de dagelijkse geïntegreerde blootstelling en dosis, 3) vaststelling van de individuele dagelijkse blootstelling, 4) gemodelleerde schatting van de vaste deeltjes (particulate matter: PM) gedeponeerde dosis in het luchtwegen, 5) bronherkenning van binnenshuis PM in een verzorgingstehuis.

Voor dit proefschrift was het belangrijk om de verblijfspatronen van de ouderen beter te begrijpen: 95 % van hun tijd werd binnenshuis doorgebracht, verdeeld over slaapkamer en huiskamer.

Hoofdstuk 2 beschrijft de karakteristieken van tien geselecteerde verzorgingstehuizen voor ouderen, en van hun 384 daarin ondergebrachte ouderen, in Lisabon en Loures, Portugal. Enige problematische verontreinigingen werden geïdentificeerd, zoals carbondioxide, vluchtige organische verbindingen, en deeltjes (PM). PM's grootste dagelijkse impactperiode was geassocieerd met opstaan s'morgens, en met specifiek gedrag en bewegingen, zoals ontbijt- en lunchtijd, in slaapkamers en huiskamers respectievelijk: tijdsgebonden concentratiepatronen maakten het mogelijk om "hot-spots" of dagelijkse "probleemmomenten" te herkennen.

Chemische karakterisering maakte het mogelijk om tot identificatie van binnenshuis- en buitenshuis emissiebronnen te komen, en om tot maatregelen te komen om deze emissies te beperken. De resultaten lieten zien dat, hoewel ouderen in dezelfde omgeving leven, hun blootstelling en geïnhaleerde dosis significant kunnen verschillen. De data laten daarbij ook zien dat een accurate bepaling van de geintegreerde blootstelling essentieel is om tot een adequate evaluatie te komen van de PM dosis-response relatie. De uitkomsten zijn belangrijk i) om de kritische gebieden binnen de verzorgingstehuizen te herkennen, ii) om de micro-milieu's te kunnen identificeren met de hoogste impact op de ouderenblootstelling en dosis, en iii) om de relevantie te herkennen van de geografische liggen van verzorgingstehuizen voor ouderen.

In Hoofdstuk 3 worden PM blootstelling en dosis berekend voor zelfstandige ouderen en ouderen in verzorgingstehuizen. Zelfstandige ouderen droegen een real-time PM-monitor gedurende vier 24h meetcampagnes, en vulden tegelijkertijd een activiteitendagboek in. Daarna werden PM gedeponeerde doses bepaald met behulp van een computerprogramma dat schattingen deed ten aanzien van transport en depositie van PM in de humane luchtwegen. In het algemeen was de gedeponeerde fractie groter in mannen dan in vrouwen

voor dezelfde activiteit, en liet een stijging zien met het activiteitsniveau. De resultaten waren belangrijk om te begrijpen waarom bepaalde gebieden binnen de verzorgingstehuizen tot hoge blootstelling aanleiding gaven, om tot effectieve maatregelen te kunnen komen, en om op deze wijze tot een verlaging van PM blootstelling en gezondheidsproblemen te kunnen komen voor deze heel gevoelige leeftijdsgroep.

Hoofdstuk 4 geeft de resultaten van de binnenshuis PM bronherkenningsstudie in een geselecteerd verzorgingstehuis.Voor deze studie werden twee bemonsteringscampagnes uitgevoerd, waarbii  $PM_{10}$ werd verzameld. De elementsamenstelling, ionen, en de organische- en elementaire koolstof werden bepaald om tot deze bronherkenning te kunnen komern. Buitenshuis PM<sub>10</sub> concentraties waren significant hoger gedurende de dag dan gedurende de nacht (p-waarde < 0.05), zowel als Ca<sup>2+</sup>, Fe, Sb en Zn. Zowel binnenshuis als buitenshuis PM<sub>10</sub> gemiddelde concentraties overschreden geen bestaande richtlijnen, en er waren geen seizoensgebonden verschillen. Het aandeel van binnenshuizige- en buitenshuizige bronnen werd bepaald via Principal Component Analyse, de uitkomsten gaven de relevantie aan voor de luchtkwaliteit, van nabijgelegen hoofdwegen en het vliegveld, op minder dan 500 m verwijderd van het verzorgingstehuis.

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### **CURRICULUM VITAE**

M. Almeida-Silva was born in 1988 and graduated in Environmental Health by Escola Superior de Tecnologia da Saúde de Lisboa (ESTeSL) in 2010. In 2010 was project assistant in CV-Dust – Atmospheric aerosol in Cape Verde region: seasonal evaluation of composition, sources and transport, project funded by Fundação para a Ciência e Tecnologia (FCT) (PTDC/AAC-CLI/100331/2008). Since 2011 is developing her PhD in TU Delft in Environmental Science: Elderly Exposure Assessment to Air Pollutants (fellow funded by FCT: BD 69700/2010). Between 2010/2012 M. Almeida-Silva performed duties and Assistant Professor in ESTeSL in Graduated Course of Environmental Health. M. Almeida-Silva published one book chapter and 22 scientifically articles (WoS), 8 as first author, in international peer-reviewed journals. Also published 5 scientifically articles (non-WoS) in international peer-reviewed journals, 16 publications at international conference proceedings with ISBN, 11 publications at international conference proceedings without ISBN, performed 11 oral presentations as author and presenter and 21 poster presentations in international conferences.

M. Almeida-Silva has 151 citations and an h-Index of 9. She also participates on the organization of 4 international conferences.

### LIST OF PUBLICATIONS

# **Book's Chapters:**

1 - Freitas, M. C., Canha, N., Martinho, M., Almeida-Silva, M., Almeida, S. M., Pegas, P., Alves, C., Pio, C., Trancoso, M., Sousa, R., Mouro, F., Contreiras, T.: Indoor Air Quality in Primary Schools, 2011. Advanced Topics in Environmental Health and Air Pollution Case Studies, Anca Maria Moldoveanu (Ed.), ISBN: 978-953-307-525-9, InTech, Available from: http://www.intechopen.com/articles/show/title/indoor-air-quality-in-primary-schools.

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- 22 Almeida-Silva, M., Almeida, S.M., Pegas, P.N., Nunes, T., Alves, C.A., Wolterbeek, H.T., 2015. Exposure and dose assessment to particle components among an elderly population, Atmospheric Environment, 102: 156-166. doi:10.1016/j.atmosenv.2014.11.063 (n.º of citations: 1).
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- 15 Almeida-Silva, M., Faria, T., Pilou, M., Housiadas, C., Wolterbeek, H.T., Almeida, S.M., 2014. Personal daily exposure to particulate matter: an elderly study. 3rd International Congress on Environmental Health, Porto, Portugal, 24 26 September 2014. ISBN: 978-989-20-5086-7.
- 14 Almeida, S.M., Almeida-Silva, M., Pinto, M., 2014. Indoor air quality in Portuguese buildings: new regulations, 3rd International Congress on Environmental Health, Porto, Portugal, 24 26 September 2014. ISBN: 978-989-20-5086-7.
- 13 Viegas, S., Almeida-Silva, M., Sabino, R., Viegas, C., 2014. Exposure to volatile organic compounds, particulate matter and fungi in a composting plant, 2014. International Symposium on Occupational Safety and Hygiene, Guimarães, Portugal, 13 -14 February 2014. ISBN 978 1-138-00144-2.
- 12 Viegas, S., Almeida-Silva, M., Sabino, R., Viegas, C., 2014. Occupational exposure to particulate matter and fungi in a composting plant case study in Portugal, 2014. International Symposium on Occupational Safety and Hygiene, Guimarães, Portugal, 13 14 February 2014. ISBN 978 1-138-00144-2.
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# **International Conference Proceedings without ISBN:**

11 - Almeida-Silva, M., Faria, T., Saraga, D., Maggos, T., Wolterbeek, H.T., Almeida, S.M, 2014. Source apportionment of indoor PM10 in elderly care center. The 13th

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- 10 Almeida-Silva, M., Pilou, M., Faria, T., Almeida, S.M., Housiadas, C., Wolterbeek, H.T., 2014. Elderly daily exposure and internal dose of particles modelling based on personal aerosol measurements, The 13th International Conference on Indoor Air Quality and Climate Indoor Air 2014, Hong Kong, 7-12 July 2014.
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- 11 Almeida-Silva, M., Wolterbeek, H.T., Almeida, S.M., 2015. Investigation on PM10 Composition and Sources Influencing Elderly Exposure. 11<sup>th</sup> International Conference on Nuclear Analytical Methods in the Life Sciences, NAMLS11, Delft, The Netherlands, 23-28 August 2015.
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- 4 Almeida-Silva, M., Almeida, S.M., Pio, C. A., Nunes, T., Cardoso, J., 2012. Elemental composition of air particulate matter in Cape Verde. European Aerosol Conference (EAC2012), Granada, Spain, 2-7 September 2012.
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- 21 Almeida-Silva, M., Lage, J., Canha, N., Silva, A.V., Almeida, S.M., 2015. The role of INAA in source apportionment studies. 11<sup>th</sup> International Conference on Nuclear Analytical Methods in the Life Sciences, NAMLS11, Delft, The Netherlands, 23-28 August 2015.
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- 19 Ramos, C.A., Canha, N., Almeida-Silva, M., Lino, J., Faria, T., Cabo Verde, S., Viegas, C., Marta, S.M., 2014. Exposure of vulnerable groups of people to bioaerossols. Trends on Environmental Microbiology and Public Health, Lisbon, 17-20 September 2014.
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- 17 Almeida, S. M., Almeida-Silva, M., Pio, C.A., Nunes, T., Cardoso, J., Cerqueira, M., Reis, M.A., Chaves, P.C., Taborda, A., 2013. Saharan dust contribution to PM10 levels and composition in Cape Verde. European Aerosol Conference 2013, Prague, Czech Republic, 1-6 September 2013.
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- 13 Almeida-Silva, M., Almeida, S.M., Pio, C.A., Nunes, T., Cardoso, J., Chaves, P.C., Taborda, A., Reis, M.A., 2013. Elemental characterization of aeolian aerossol from Cape

- Verde by INAA and PIXE. European Geosciences Union (EGU2013), Vienna, Austria, 7 12 April 2013.
- 12 Almeida-Silva, M., Almeida, S.M., Pio, C.A., Nunes, T., Cardoso, J., Chaves, P.C., Taborda, A., Reis, M.A., 2013. Elemental characterization of aeolian aerossol from Cape Verde by INAA and PIXE. European Geosciences Union (EGU2013), Vienna, Austria, 7-12 April 2013.
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- 8 Canha, N., Almeida-Silva, M., Almeida, S.M., Freitas, M.C., Wolterbeek, H. Th., 2012. Chemical composition of total particulate matter from indoor school environments. Urban Environmental Pollution, Amsterdam, The Netherlands, 17-20 June 2012.
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- 5 Canha, N., Almeida-Silva, M., Freitas, M.C., Almeida, S.M., Wolterbeek, H. Th., 2011. Lichens as biomonitors at indoor environments of primary schools, Thirteenth International Conference on Modern Trends in Activation Analysis, College Station, Texas, USA, 13-18 March 2011.
- 4 Canha, N., Freitas, M. C., Almeida, S. M., Almeida, M., Ribeiro, M., Cabo Verde, S., 2010. Indoor air quality parameters at primary schools in Lisbon by passive methods, 2010. Air Pollution and Health, San Diego, California, EUA, 22-26 March 2010.
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# ELDERLY EXPOSURE TO AIR POLLUTANTS

Measuring, assessing and modelling