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Ultrafine Particles In Children's Home Environments

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Abstract

Ultrafine particles (UFPs) have an aerodynamic diameter lower than 100 nm. Due to their large surface area they are more reactive and toxic and can enter the human body through the skin, respiratory and gastrointestinal tract. Human exposure to UFPs in homes is associated with health risks such as cardiovascular disease and/or respiratory problems. These risks are heightened by the long time that people spend indoors. Children have higher exposures than seen in adults due to their relative higher amount of air inhalation and their growing tissue and organs.

The purpose of this work was to evaluate UFP number concentrations in different children's home environments, mainly in children bedrooms and living rooms.

UFP number concentrations were sampled in 14 urban homes located in Porto Metropolitan Area, Portugal. UFP were sampled concurrently both indoors and outdoors during Saturdays, for a period of 8 hours between January and May of 2015.

Additionally, two types of questionnaires were filled. One focused on the occupancy/activities held in the rooms where sampling equipment took place; another was dedicated to external and internal building description (e.g., location, materials, ventilation, heating, cleaning activities, sources of allergens).

The overall mean of UFP levels in bedrooms of all 14 homes was 2.51×10^4 particle cm^{-3} and 2.86×10^4 particle cm^{-3} for living rooms. Highest UFP levels were identified in homes with smoking occupants and where pets were present. The lowest values measured were in non-smoking homes where room volumes were significantly bigger. Regarding outdoor ambient concentration levels of UFP, the higher UFP mean number concentration were observed in a home located close to a road with high vehicle density (2.66×10^4 particle cm^{-3}).

I/O ratios were bigger than 1 for living rooms in all homes, indicating that emissions from indoor sources were the main contributor to indoor UFP levels. On the other hand I/O ratios of UFP measured in bedrooms were below 1 for the majority of the homes which suggests that the activities conducted in these microenvironments had lower impacts on the respective indoor UFP levels. These results are somewhat assuring considering the long period of time that children spent in their bedrooms.

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Nomenclature

Abbreviations

CCN	Cloud Condensation Nuclei
DNA	Deoxyribonucleic Acid
ECRHS	European Community Respiratory Health Survey
EU	European Union
I/O	Indoor/Outdoor Ratio
PM	Particulate Matter
$PM_{2.5}$	Particulate matter which passes through a size selective inlet with a 50% efficiency cut-off at 2.5 µm aerodynamic diameter
PM_{10}	Particulate matter which passes through a size-selective inlet with a 50% efficiency cut-off at 10 µm aerodynamic diameter
PNC	Particle Number Concentration
PMA	Porto Metropolitan Area
SINPHONIE	Schools Indoor Pollution and Health: Observatory Network in Europe
UFP	Ultrafine Particle
U.S.EPA	United States Environmental Protection Agency
VCI	Via de Cintura Interna
VOC's	Volatile Organic Compounds
WHO	World Health Organization

Elements and Compounds

Al	Aluminum

Ca	Calcium
Cd	Cadmium
CO	Carbon Monoxide
Cl	Chloride
Cr	Chromium
Cu	Copper
Hg	Mercury
Mn	Manganese
Na	Sodium
NH_4	Ammonium
Ni	Nickel
NO_3	Nitrate
NO_2	Nitrogen Dioxide
NO_x	Nitrogen Oxides
O_3	Ozone
Pb	Lead
PO_4^{2-}	Phosphate
Si	Silicon
SO_4^{2-}	Sulphate
SO_2	Sulfur Dioxide
Ti	Titanium
Zn	Zinc

1. Introduction

1.1 Relevance and motivation

Air pollution continues to be a matter of great discussion and importance due to its significant threat to human health, leading to lower life expectancy around the world (WHO, 2006). Airborne particulate matter (PM) is one of the pollutants that causes an array of adverse health effects. In the past years, there has been an increased interest in the research on sources, concentration levels and human exposure to airborne particles because of a wide range of adverse health outcomes on the human cardiovascular and respiratory systems. Susceptibility to the air pollution varies with health and age being the elderly and the children the most susceptible groups.

Ultrafine particles (UFPs), namely particles with a diameter less than 100 nm, have a high deposition fraction, meaning that UFP large surface area increases the potencial to enter the lungs and translocating to other parts of the body, potentially carrying toxic compounds with them (Heal et al., 2012). UFPs are ubiquitous in urban air where their major source is motor traffic (Kumar et al., 2014).

Indoor sources of UFPs mainly include tobacco smoke (including second hand smoke), household air pollution from combustion of solid fuels and ambient particulate matter (He et al., 2004). The relevance of studying indoor air quality is due to the fact that people spend most of their time indoors and the nature of emissions from indoor sources is different from that of the outdoor ones; composition and toxicity of UFPs may also be different from those of the outdoor particles (Morawska et al., 2009). Therefore, it is important to evaluate the UFP dose-response relationship which requires accurate measurements of personal exposure levels (Buonanno et al., 2014).

The purpose of this thesis is to obtain information of indoor sources of particles, especially focusing on indoor UFP levels in home environments situated in the Porto Metropolitan area, Portugal. These results may allow further understanding of the behaviour and dynamics of this pollutant in indoor home environments and its risks of exposure for children living in these environments. In addition, the obtained data provides information about the potential indoor sources of these particles. This study also gives an insight of spatial distribution of particles within the different home microenvironments as well as continuous information on ultrafine particle concentration during different periods of day. In addition, the results of this study were supported by detailed characterization of homes and information about time activity patterns of home occupants, which allowed further analysis of the relationship with UFP concentration variations.

The study of UFPs is particularly relevant for young children who are one of the most susceptible subgroups of population to health effects air pollution because their bodies are still developing, and they breathe at a higher volumetric rate per body mass than adults (Laiman et al., 2014). Therefore, further information on child exposure to indoor UFP may provide a contribution for the improvement of child life quality and gives background information for further studies.

1.2 Objectives

The main aim of this work was to evaluate indoor ultrafine particles in home environments. The specific objectives of this work were:

- to evaluate indoor ultrafine particle number concentrations in fourteen Portuguese homes, in comparison with other international studies;

- to evaluate indoor emission sources and the effect on ultrafine particle number concentration as well as potential outdoor sources.

1.3 Thesis outline

This thesis is divided into 5 chapters. Each associated with the following content.

Chapter 1 (present chapter) explains the relevance and motivation of this work and also explains how the thesis is structured.

Chapter 2 (state of the art) gives an overview of the problematic of air pollution and theoretical information about particulate matter. Because this work is focused on the ultrafine particles, this chapter also describes in detail regarding its properties, sources, formation processes, composition and health effects. At the end a brief description of the legislation applicable to indoor air pollution is introduced.

Chapter 3 provides the materials and methods used for this work. It describes the details of the sampling homes and sampling procedures for collection of outdoor and indoor pollutants, including equipment characteristics. The data treatment and statistical analyses made in this work are also described.

Chapter 4 contains the results and the discussion of the work. It consists of the analysis of the obtained indoor and outdoor ultrafine particle number concentrations and their comparison. Finally, a brief comparison with similar published studies is given.

In the last chapter, **Chapter 5**, conclusions of the thesis and suggestions for future studies are presented.

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2. State of the art

2.1 Air Pollution

Industrial Revolution marked a shift from a society that relied on wave and wind power, and solar energy for the production of food and goods, to an industrial and urban society, which became to rely on the burning of fossil fuels. Industrialization was characterized by powered, special-purpose machinery, factories and mass production. The development of coal mining and the use of steam power generated from coal was the central event of the nineteenth century, releasing the first emissions of carbon dioxide and other gases which led to the declining of air quality in many cities. However, air pollution only became a health concern when public started to be threatened by the rise of death rates. That awareness was important but lack of effective policies and poor enforcement drive allowed many industries to bypass laws made by pollution control board which resulted in mass scale pollution that affected lives of many people.

Today, due to an unstoppable urban development, air pollution continues to affect population on a daily basis and therefore is a major concern topic of studies and research. Ambient air in urban areas is more polluted than overall atmosphere due to the high density of human population and their activities in urban areas. This concentration of population and their activities produces a higher rate of air pollutants compared to less-developed areas and natural environments (Mabahwi et al., 2014).

The increase of sophisticated measurements of atmospheric composition and epidemiological methods reveals associations between a range of air pollutants (and its lower concentrations) and adverse health outcomes (Heal et al., 2012). The main pollutants released directly into the atmosphere in their unmodified forms in sufficient quantities to pose a health risk concern include particulate matter (PM), carbon monoxide (CO), ozone (O_3) and nitrogen and sulfur dioxide (NO_2 and SO_2).

Studies have been made to evaluate the effects of air pollution on morbidity and mortality. According to WHO assessment of the burden of disease due to air pollution; more than 2 million premature deaths each year can be attributed to the effects of both urban outdoor and indoor air pollution (WHO, 2006). Some of the consequences of air pollution on public health have been the subject of epidemiology and laboratory studies and include the major respiratory problems such as bronchitis, emphysema and asthma, cardiovascular diseases and lung cancer (Heal et al., 2012; Mabahwi et al., 2014).

2.2 Particulate Matter

Particulate matter is one of the most important air pollutants mainly because it can contribute to the prevalence of chronic respiratory diseases like asthma. PM is a mixture of both solid and liquid phase material suspended in the air and may have a diverse chemical and physical composition depending on its source (Branco et al., 2014).

Particulate matter has a complex nature and a highly variable size, ranging between molecular dimensions to the sizes that are distinguishable with the naked eye which represents a problem in the choice and availability of instrumental techniques. The characteristics of PM are wide and only some of them can be measured simultaneously. Their sources are still to be clearly identified, they can be from outdoor origin which has penetrated indoors or it can be generated by indoor sources being the nature of emissions from indoor sources different from that of the outdoor sources, producing ultrafine particles (i.e. particles smaller than 100 nm) with different composition and toxicity (Morawska et al., 2013; Morawska et al., 2009).

PM can be emitted directly into the atmosphere as particles (primary particles) or formed within the atmosphere itself from nucleation and condensation reactions of gas-phase species (secondary particles) and then continues its chemical and physical transformation in the atmosphere (Heal et al., 2012; WHO, 2006).

The exact composition of these particles varies with their size range, location and prevailing meteorology. However, the major chemical constituents of airborne PM are known to be sulphate (SO_4^{2-}), nitrate (NO_3^-), ammonium (NH_4^+), sodium (Na) and chloride (Cl) ions, elemental and organic carbon, mineral material and water. Other minor and trace components are phosphate (PO_4^{3-}) and other elements particularly metals such as Pb, Cd, Hg, Ni, Cr, Zn, Mn, emitted from a wide range of metallurgical industries, from vehicle engine, brake and tyre wear, and during combustion of impure fuel and fuel and lubricating oil additives (Heal et al., 2012).

The potential of particles to cause damage varies with the size and other physical characteristics, and chemical composition (WHO, 2006). It has been shown that the fine PM fraction is the one that most affect human health. That is why recently measurements of total suspended PM (PM_{Total}) have been replaced by finer particles, ultrafine particles (Branco et al., 2014).

Particles are classified by their aerodynamic properties because these strongly influence particle lifetime in the atmosphere, the spatial extent of their influence and clearance pathways within the respiratory tract (Heal et al., 2012; WHO, 2006). Regarding the physical characteristics, PM aerodynamic diameter ranges from few nanometers (nm) to several micrometers (µm) (Heal et al., 2012). In that regard, particulate matter can be divided into PM_{10}. i.e. particles below 10 µm in aerodynamic diameter and $PM_{2.5}$ which includes particles with aerodynamic diameter less than 2.5 µm. PM_{10} includes inhalable particles that are sufficiently small to penetrate the thoracic region and $PM_{2.5,}$ which is fraction that is included in $PM_{10,}$ has a high probability of deposition in the smaller conducting airways and alveoli (WHO, 2006).

The distribution of ambient particles as a function of particle size, whether in urban or non-urban environments, is characterized by three modes: nucleation or ultrafine mode, fine mode and coarse mode. These size fraction modes differ in their overall contributions to airborne particle mass and in their origins, physical characteristics and chemical composition (WHO, 2006). *Figure 1* demonstrates how the distribution of atmospheric particles varies with its aerodynamic diameter. The overwhelming majority of particles contributing to a total mass concentration have diameters larger than 0.1 µm. This, among the differences in properties, leads to the separate treatment of UFPs and larger particles (Nazaroff, 2004).

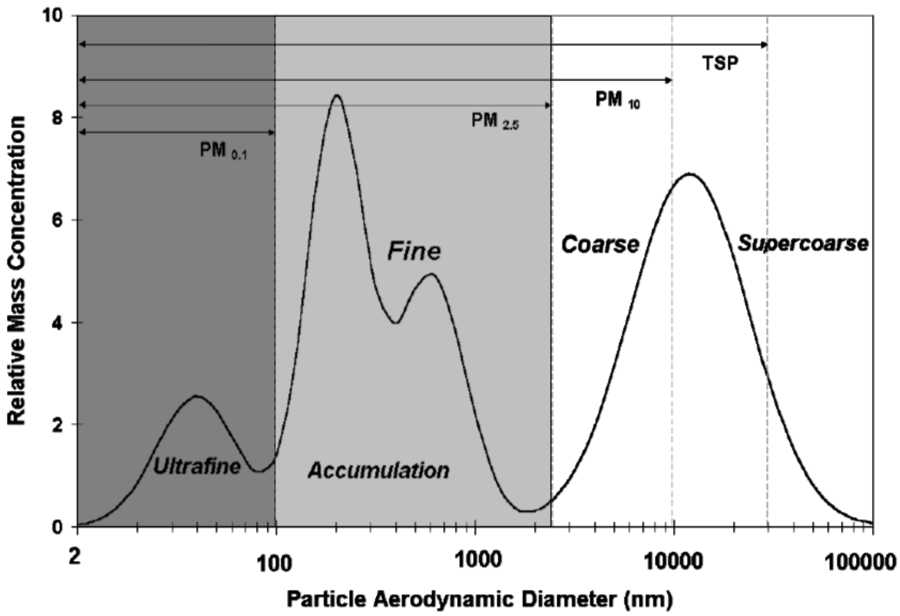

Figure 1 - Schematic representation of the size distribution of atmospheric particles adapted from Slezakova et al., 2013)

2.2.1 Indoor Air Quality and Exposure

The scientific evidence has shown that the air in home environments and other buildings can be more seriously polluted than the outdoor air even in the largest and most industrialized cities (Branco et al., 2014). The majority of the people, especially susceptible subgroups of population, spend most of their time in indoor environments such as homes, day-care facilities, offices, shopping malls, among others, and thus can be exposed to the effects of indoor air pollution. Such groups include the young, the elderly, and the chronically ill, especially those suffering from respiratory or cardiovascular disease (Agência Portuguesa do Ambiente, 2015; United States Environmental Protection Agency, 2014).

Given that most people spend the majority of their time indoors and since indoor peak concentrations can be several times higher and more frequent than outdoor concentrations, the

consequences of the respective exposure range from insignificant to fatal and depend on the type of various parameters such as pollutant present, type of indoor environment, duration of time spent, age, gender, susceptibility. and many other factors (Morawska et al., 2013).

Concerning particles, both indoor and outdoor sources contribute to and affect the concentration and composition of particles in indoor air (He et al., 2004). Indoor particles are a mix of ambient particles that have infiltrated indoors. particles emitted indoors and particles formed indoors through reactions of gas-phase precursors emitted both indoors and outdoors (Morawska et al., 2013). The composition and toxicity of indoor particles is very complex and there is a strong need to study its characteristics and health effects.

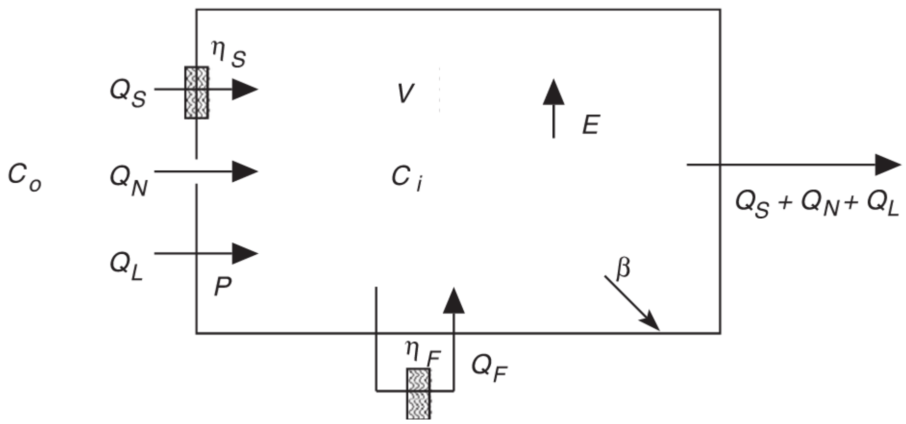

Figure 2 - Schematic representation of indoor particle dynamic processes (adapted from Nazaroff, 2004).

Theoretically, indoor particle concentration is a function of a number of factors, the most important of which are the generation rate of particles indoor, the outdoor particle concentration, air exchange rate, particle penetration efficiency from the outdoor to the indoor environment, and the particle deposition rate on indoor surfaces (Morawska et al., 2001).

Figure 2 represents an illustration of the processes that occur inside an indoor environment that affect indoor particle levels. Inside the building there may be one or more emission sources, represented by the letter E. A deposition of some particles onto room surfaces, β may also occur. Gravitational settling of particles depend highly on their size hence deposition rate is lower for smaller particles. Among the factors that can increase particle deposition are a higher airflow and increased indoor surface area (Nazaroff, 2004).

Outdoor air, which contains a certain concentration of particles, C_0, enters the compartment through ventilation by three different ways: by natural ventilation, Q_N, by mechanical supply, Q_S, and by leakage (infiltration), Q_L. Mechanical ventilation systems are usually equipped with fibrous filters that retain some of the particles which is why it is considered their removal efficiency, η. In the illustration there are represented two flow paths that passes through filters with a flow rate of Q_R and Q_F and efficiency of η_R and η_F. Some of the outdoor air particles also enter indoors by penetrating through small cracks or gaps in the building envelope, P. Penetration, source emission, and deposition rates all vary depending on particle size. The total indoor concentration of particles is represented by C_i and V represents the interior volume of the compartment in study (Nazaroff, 2004).

The term *exposure* implies that there is a hazardous component that. in contact with humans possesses a risk to their health. It is a product of the pollutant concentration and the time over which a person is in contact with that pollutant. In terms of duration, exposure to elevated concentrations can be short-termed or during lifetime. In this context, exposure assessment includes the identification of indoor sources of pollution, the characterization of indoor air pollutants and contributing factors such as concentration, duration, frequency, distribution route, time frame, and geography and also the modelling of exposure for different occupancy and lifestyle scenarios. Quantification of personal exposure can be supported by direct personal exposure measurements (which are the most accurate), by measuring microenvironmental concentrations of a pollutant and time spent in these microenvironments, and by collecting personal activity information. All of this can be done through measurements, surveys, questionnaires or other methods (Morawska et al., 2013).

Specifically, assessment of human exposure to indoor particulate matter can be limited due to three main reasons: (i) the definition of exposure is often unclear; (ii) particulate matter characteristics are complex; and (iii) the origin of the particulate matter is not clear (Morawska et al., 2013; WHO, 2006).

In general, there is considerably less information available on indoor ultrafine particles, and the systematic quantification of various indoor source emission rates of these particles, including particle formation mechanisms, particle concentration levels, and human exposure, are still to be investigated (Morawska et al., 2009). While the past decade has seen an increase in the body of literature published on this topic, there are still major challenges to be addressed, to fully understand and quantify the magnitude of both individual and population exposure to UFP air pollution in different types of indoor microenvironments.

2.2.2 Coarse Mode

The coarse mode particles are the particles with a higher size, i.e. particles with aerodynamic diameters larger than 2.5 μm. Because of their large size, coarse particles readily settle out or impact on surface, so their lifetime in the atmosphere is short. These particles are mainly primary particles once their major source is mechanical abrasion processes, meaning that they are produced by the break-up of larger solid particles. However, biological sources may also contribute to this mode. Because of the different processes occurring in the atmosphere, these classes of particles change dynamically (Friend et al., 2012). Coarse particles include earth's crustal materials such as wind-blown dust from agricultural processes, uncovered soil, unpaved roads or mining operations. In urban areas these particles contain resuspended dust from roads and industrial activities and biological material such as pollen grains and bacterial fragments (WHO, 2006).

The coarse mode can further be subdivided into supercoarse and coarse particles. The coarse particles have an aerodynamic diameter between 25000 nm (i.e. 25 μm) and 10000 nm (i.e. 10 μm) that refers to the fraction of particles that are produced by mechanical processes. This fraction deposits in the upper airways of the human respiratory system and is eliminated from human body through the nose or by coughing or swallowing. Supercoarse particles have a diameter bigger than 10 µm and they are too big to enter human respiratory system, representing no harm to human health. However, in some studies this fraction is also measured and evaluated due to its possible environmental impacts (Slezakova et al., 2013).

2.2.3 Fine Mode

Particles smaller than 2.5 µm in aerodynamic diameter constitute the fine mode. This fraction also reaches the alveolar region of the human respiratory system.

Fine mode particles are subdivided into accumulation and nuclei modes based on the formation mechanisms. Accumulation mode consists of particles with a diameter between 100 and 1000 nm. These particles are generated by anthropogenic sources (combustion processes) but can be formed by natural processes (Slezakova et al., 2013). Natural processes include nucleation-condensation of low-vapour-pressure substances formed by high-temperature vaporization or by chemical reactions in the atmosphere. These small particles tend to accumulate by coagulation or by condensation of gas or vapour molecules on the surface of existing particles. Coagulation is more efficient for large number of particles, while condensation is most effective for large surface areas (WHO, 2006). Accumulation mode particles are comparable with the wavelengths of visible light which contributes to the anthropogenic visibility impairment problem in urban areas (Slezakova et al., 2013).

2.3 Nuclei Mode – Ultrafine Particles

2.3.1 Properties

Nuclei mode particles or ultrafine particles (UFPs) have an aerodynamic diameter lower than 100 nm. This fraction of particles is commonly measured and expressed in terms of number concentrations of particles per unit of volume of air and not in terms of mass concentration like larger particles (Slezakova et al., 2013).

Their small size allows them to entry into all parts of the lung including the alveoli as well as other human cells of various tissue types (Kearney et al., 2011) .

UFPs are found in high number concentrations near their sources. Due to their small size and large surface area. they are highly chemically reactive and are still going through chemical reactions or modification processes after their release to atmosphere (Slezakova et al., 2013).

2.3.2 Sources

There are numerous sources of particles related to both human activities as well as natural sources. Although specific source impacts differ between regions, combined analyses suggest that in developed countries more than two thirds of UFPs is traceable to anthropogenic sources (WHO, 2006).

Quantitative assessment of indoor source emission characteristics in real situations is a complex task, and therefore only qualitative information about the contribution of many indoor particle sources, or about indoor ranges of concentration levels as a result of operation of the sources is available. However, a large number of indoor particle sources have been identified and emissions from these sources investigated by many studies. The most significant sources include tobacco smoking, cooking, kerosene heating and wood burning (He et al., 2004). Other sources or human activities that contribute to elevated levels of indoor particles include dusting and vacuuming, showering, operation of humidifiers, candles or incense burning, using laser printers, etc. (Géhin et al., 2008; Laiman et al., 2014). Combustion processes are the main indoor sources of smaller particles, with the vast majority of them in the submicrometer range, containing a host of organic and inorganic material. (Abt et al., 2000; He et al., 2004).

2.3.3 Processes

Ultrafine particles are produced by combustion and atmospheric processes (Wallace, 2006). They can be produced by the condensation of metals or organic compounds that are vaporized in high-temperature combustion processes, and by the condensation of gases that have been converted in atmospheric reactions to low-vapour-pressure substances (nucleation). The main precursor gases are sulfur dioxide (SO_2), nitrogen oxides (NO_x), ammonia (NH_3) and volatile organic compounds (VOCs) (WHO, 2006).

2.3.4 Composition

There is still a lack of knowledge about the composition of ultrafine particles. especially because it can be influenced by their sources and formations and also depends on geographical and meteorological parameters (Slezakova et al., 2013). However it is known that ultrafine particles are composed of both primary and secondary particulate matter. The primary fraction is typically the dominating concentration and is emitted directly from the emission source, often includes aggregates of smaller particles (Kim et al., 2001). The secondary component is composed of particulate matter formed in the atmosphere, including sulfuric acid and sulfates. and organic reaction products of low volatility (Kim et al., 2001). This size fraction is partially distinguishable from other directly anthropogenic sources. These changes involve photoreactions of oxides of nitrogen (NOx), and sulfur dioxide (SO_2), both of which are products of combustion. There is also a component of secondary particle chemistry that result in production of ammonium sulphates, nitrates, and chlorides, but these materials are thought to have less toxicological significance (Donaldson et al., 2003). Crustal minerals also contribute some fraction to the amount of ultrafine particles in the environment, which can be identified through the presence of metals such as Si, Al, Ti and Ca. There appears to be a strong correlation with UFP's and NO, NO_2, CO, Zn, and Cu, which appear to reflect motor vehicle traffic (Cyrys et al., 2003).

2.3.5 Environmental Effects

Healthy environment has a strong connection to human health. Ultrafine particles play a role in climate change as the particles directly affect climate by enhancing the scattering and absorption of solar radiation, therefore altering the amount of solar radiation reaching the Earth's surface. Aerosols can greatly contribute to the number of cloud condensation nuclei (CCN) through growth by condensation, affecting cloud properties and the hydrological cycle and therefore having a great impact on the rise of global climate light scattering from aerosols leads to visibility degradation. They can also act as sinks for reactive species since their exposed surfaces can catalyze heterogeneous reactions (Posner and Pandis, 2015).

2.3.6 Health Effects

Ultrafine particles can enter the human body through the skin, respiratory and gastrointestinal tract. Due to their large surface area they are more reactive and toxic (Slezakova et al., 2013). Ultrafine deposition in human lungs results in inflammation, impairment of phagocytosis and thrombosis. Ultrafine particles can become deposited in the lungs and trigger asthma attacks or it can be transported by the blood to the central nervous system or to other organs such the liver (Slezakova et al., 2013). Animal and human studies have shown that ultrafine particles might be even more strongly associated with effects on various health endpoints than fine particles, including oxidative damage to DNA repair mechanisms and total and cardio-respiratory mortality (Kearney et al., 2011). More recently, transportation of ultrafine particles to the brain via the olfactory nerve has been demonstrated (Wallace, 2006). Personal exposure to UFP led to significantly worsened heart rate variability for young adults and elderly adults with impaired lung function (Wallace, 2006).

People that are at most risk of exposure to indoor particles include infants, children and teens, people over 65 years old, people with lung disease (asthma, chronic pulmonary disease, chronic bronchitis and emphysema), people with heart disease or diabetes (Mabahwi et al., 2014). Children's not fully developed immune system and lungs, their relative higher amount of air inhalation and their growing tissue and organs, which together raise the possibility of higher exposures than seen in adults (Branco et al., 2014).

So far there is no quantitative information about concentration-response functions for UFPs that can be used in health impact assessment and the epidemiological studies on the health effects of these particles are very few (Slezakova et al., 2013).

2.4 Legislation

The European Union introduced the Directive 2010/31/EU in order to improve energy performance of buildings for EU member states. This directive takes into account climatic and local conditions as well as indoor climate environment and cost-effectiveness (EU Directive, 2010).

This Directive was approved and transposed to Portuguese legislation in Decreto-Lei 118/2013 which promotes the improvement of energetic performance of buildings facing the goals and challenges agreed by the member states to 2020 (Decreto-Lei n.º 118/2013).

The Portuguese limits for levels of indoor air pollutants are established in Portaria 353-A.2013. It establishes the minimum values of new airflow by space, as well as thresholds of protection and reference conditions for indoor air pollutants of new services and commercial buildings, and the respective evaluation methodology (Portaria n.º 353-A/2013). The proposed thresholds of protection are shown in *Table 1*.

Table 1 - Portuguese threshold limits for indoor pollutants and tolerance margin (Portaria n.º 353-A/2013).

Pollutants	Units	Protection threshold	Tolerance margin (%)
Suspended particles (PM_{10})	$\mu g\ m^{-3}$	50	100
Suspended particles ($PM_{2.5}$)	$\mu g\ m^{-3}$	25	100
Total volatile organic compounds (VOC's)	$\mu g\ m^{-3}$	600	100
Carbon monoxide (CO)	$mg\ m^{-3}$ ppmv	10 9	-
Formaldehyde (CH_2O)	$\mu g\ m^{-3}$ ppmv	100 0.08	-
Carbon dioxide (CO_2)	$mg\ m^{-3}$ ppmv	2250 1250	30
Radon	$Bq\ m^{-3}$	400	-

These limits are established as an average of 8 hours measurements and are applied at 20 °C and 1 atm. They also are applied to buildings without mechanical ventilation (Portaria n.º 353-A/2013).

In the previous legislation based on the Directive 2002/91/CE, $PM_{2.5}$ was not distinguished from PM_{10} and the measurement was based on an average of 5 minutes sampling. In that view, the current legislation is more stringent than the previous one.

Concerning the UFP, the regulatory aspect has not been addressed yet and there are no limits or thresholds proposals. The main problem relies on the difficulty to choose a metric that would be most adequate since a wide range of these particles' characteristics influence human exposure. At national levels air quality agencies should be encouraged to integrate ultrafine particle measurements in their monitoring networks to provide comprehensive data and information necessary to correctly address regulatory aspects of atmospheric UFPs in order to prevent public exposures (Slezakova et al., 2013).

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3. Materials and methods

3.1 Characterization of the Study Region

The study measurements were performed in homes located in the Porto Metropolitan Area (PMA).

The city of Porto is one of the oldest European cities and the second largest city in Portugal (after Lisbon). It is located in the North of Portugal along the Douro river estuary. PMA represents 2.58 % of national territory and 9.58 % of the north region. It includes 17 municipalities and has an extent of 2.040 km^2 with a resident population of about 1.7 million inhabitants and a population density of 862 inhabitants per square kilometre. The majority of the population has an age between 15 and 64 years-old (AMPorto, 2015).

PMA climate has two main influences: the proximity to the Atlantic Ocean and its mountainous eastern limitation. The temperature is mild, however its relative humidity is high and the rain abundant. The annual average temperature ranges between 13 and 15 °C. The total annual precipitation varies between 1000 and 1200 mm, nevertheless long periods with mild temperatures and sunny days are frequent even during the rainiest months.

Analytical results confirm that historical monuments in urban areas act as passive repositories for air pollutants present in the surrounding atmosphere. Despite its cultural values, the façades of monuments and historical buildings of Porto have been deteriorating, showing intense loss of materials (Slezakova et al., 2011). However, traffic emissions are considered to be the main source of air pollution on the metropolitan area following the high industrial activity density (Slezakova et al., 2011).

3.2 Homes Characterization

The characterization of indoor air quality of Portuguese homes was part of a large study identified as ARIA – How indoor air quality can affect children allergies and asthma (PTDC/DTP-SAP/1522/2012). Fourteen houses (randomly identified as C07–C55) were chosen for measurements of the ultrafine particle number concentrations. All of these homes were located in Porto Metropolitan Area, specifically in municipality of Porto, only one house was situated in municipality Gondomar. The main construction characteristics of the buildings of 14 homes are presented in *Table 2*.

Table 2 - Buildings main characteristics.

Building Characteristic	***n (%)***
Location	
City Center	6 (42.9)
Commercial/Residential Area	3 (21.4)
Suburban With Large Gardens	2 (14.3)
Town With or Without Small Gardens	2 (14.3)
Industrial/Residential Area	1 (7.14)
Year of construction	
40's	1 (7.14)
60's	2 (14.3)
90's	6 (42.9)
After 2000	5 (35.7)
Building Type	
Apartment	12 (85.7)
Semi-detached house	2 (14.3)
Main buildings materials	
Hollow Brick- Masonry	11 (78.6)
Other	2 (14.3)
No information	1 (7.14)

Number of floors	
0	1 (7.14)
1	1 (7.14)
2	1 (7.14)
3	1 (7.14)
4	5 (35.7)
> 4	5 (35.7)
Certification	
Yes	1 (7.14)
No	13 (92.9)

n = number of homes

Figure 3 shows the geographical distribution of all fourteen homes.

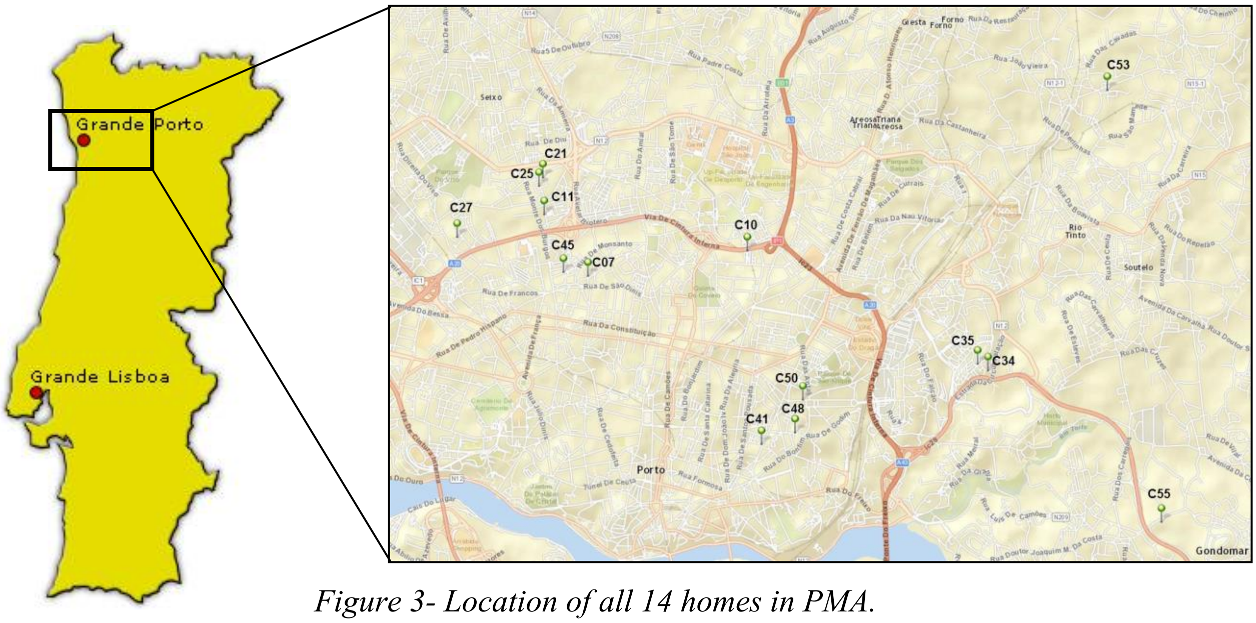

Figure 3- Location of all 14 homes in PMA.

3.3 Micro-environments

The studied indoor microenvironments typically included children bedrooms and another division of the home, most commonly the living room. If possible, UFP were measured concurrently outdoors. In some homes not all of these microenvironments could be included due to the lack of equipment available or errors caused in the measurement or due to the prohibition or unintentional neglect of the parents.

In order to better understand the fate and behaviour of indoor particles, a detailed characterization of each micro-environment was made.

3.3.1 Bedrooms

The sampling campaign of UFP was performed in the children bedrooms of all 14 homes. In every house the ventilation was naturally made by the opening of windows and doors.

The main characteristics that may influence ultrafine particle number concentrations are classified in the following table (*Table 3*).

3.3.2 Living Rooms

Whenever it was possible, UFP number concentrations were also measured in the living rooms. The ventilation was naturally made in all of the 10 living rooms studied.

Table 4 presents the most important characteristics of the studied living rooms.

Table 3 - Main characteristics of studied children bedrooms.

Characteristics	C07	C10	C11	C21	C25	C27	C34	C35	C41	C45	C48	C50	C53	C55
Volume (m^3)	32.9	31.8	29.3	35.6	23.9	25.3	23.1	28.1	29.2	33.6	33.9	34.2	38.4	34.1
Floor Area (m^2)	12.7	15.1	11.4	14.6	9.2	10.2	9.4	11.5	11.2	13.6	13.8	12.1	14.6	12.8
Openable Windows (%)	50	50	50	50	50	50	50	50	100	50	50	25	50	50
Average Temperature (°C)	17	-	20	18	18	19	18	17	20	18	20	21	23	-
Average Relative Humidity (%)	71	-	51	56	72	53	54	34	44	56	54	52	60	-
Heating System														
Hot Water Radiators/Convectors			√	√										
Electrical Radiators/Convectors		√				√								
Both	√													
Warm Air Flow										√				
None					√		√	√	√		√	√	√	√
Smoking allowed	No	Yes	No	No	No	No	No	No	No	No	No	No	No	No
Pets	No	Yes	No	No	Yes	Yes	No	Yes	No	No	No	Yes	Yes	Yes
Number of permanent room occupants	1	2	2	2	1	1	1	2	2	1	1	1	1	1
Floor Coverings														
Wood - parquet	√			√	√	√	√		√			√	√	√
Stone Tiles		√												
Acoustic Flooring			√							√	√			
Ceramic Tiles								√						
Electronic Equipment														
TV	1	1	1	1			1	1			1	1	1	1
Computer	1		1	2	1	1				1		1	1	
Printer				1		1							1	
Photocopier													1	
Audiotape						1			1					
Other electronic equipment		1		1							1			1
Visible Mould	No	No	No	No	No	No	No	No	No	No	No	Yes	No	Yes

"-" - not available

Table 4 - Main characteristics of studied living rooms.

Characteristics	C07	C10	C11	C21	C25	C27	C34	C41	C45	C48
Floor Area (m^2)	-	5-10	27	20-25	10	20	15	-	39	21
Openable Windows (%)	50	50	50	50	50	50	50	100	50	50
Heating System										
Hot Water Radiators/Convectors				√						√
Electrical Radiators/Convectors					√	√				
Both	√		√							
Warm Air Flow									√	
Closed Fireplace		√	√							
None							√	√		
Smoking allowed	No	Yes	No	No	Yes	Yes	No	No	No	No
Pets	No	Yes	No	No	Yes	No	No	No	No	No
Electronic Equipment										
TV	1	1	1	1		1		1	1	1
Computer			1	1				1		1
Printer				1					1	1
Photocopier										1
Audiotape								1		
Other electronic equipment			2							
Dehumidifier	No	No	No	No	Yes	No	Yes	No	No	No
Air Freshner	No	No	No	No	Yes	Yes	No	No	No	No
Visible Mould	No	No	No	No	Yes	No	No	No	No	No

“-“ - not available

3.3.3 Kitchen

Among the homes that were analyzed in this study, only kitchen of one home (C25) was included for measurements of UFP. This home, a house located in a commercial/residential area, had a busy road and a highway nearby; an industrial factory was situated approximately within a distance of 1 km. The total number of occupants of this house was four people, some of them who smoked inside the house, particularly in the kitchen.

The kitchen estimate area is between 5 and 10 m^2 and has windows that permits natural ventilation. Ultrafine particle number, CO_2, temperature, and relative humidity were the parameters measured for this compartment.

3.3.4 Outdoors

For comparison, ultrafine particle number concentrations were also measured in the outdoor air, however such monitoring was not possible in all homes namely in C25, C34, C35, C48, C50 and C53 due to the lack of terraces or outdoor places to install the equipment. All homes in which outdoor measurements of UFP were conducted were located in an urban environment.

3.4 Questionnaires

To collect all the information possible from every home analyzed two types of tools were used. One of the questionnaires focused on home related activities. The second one compiled information about the building and home physical characteristics (such as surface area. types of surfaces. flooring. type of ventilation and heating systems, etc.), possible sources of allergens, and the major types of children activities.

The two types of questionnaires were based on previous versions developed within European projects like SINPHONIE and ECRHS II - indoor questionnaire, indoor air quality in schools - federal environment agency, U.S.EPA's guidance documents, the Canadian centre for occupational health and safety guidance data, University of Missouri Extension - Building

Strong Families, Healthy Home Handouts and Utah State University - how to purchase a healthy home - Indoor Air Quality Assessment Checklist.

Both questionnaires are shown in the Appendix of this thesis.

Parents' diary

The parents' diary was focused on the occupancy/activities held in the rooms where sampling equipment was placed.

This questionnaire was given out to the parents or a responsible adult, who then registered the time (and duration) of a specific activities/sources that were conducted/used in the evaluated compartments of the house, in order to cross-reference them with the ultrafine particle number concentration levels.

House Characteristics Checklist

A house checklist was dedicated to external and internal building description (location, materials, ventilation, heating, cleaning activities, sources of allergens). It was filled by a researcher of the study project.

The main objective of this checklist was to collect as many information as possible, with more details and with the focus on a better understanding of all the factors that may influence the levels of the pollutant measured.

3.5 Sample Collection

The measurements were conducted between January and May of 2015, in a total of 14 homes. All measurements were performed during Saturdays, when parents allowed access to their homes, in a period of 8 h during daytime.

For continuous real time monitoring of ultrafine particle number concentration TSI P-Trak™ Model 8525 (*Figure 4*) was used.

Figure 4 - TSI P-Trak sampler (Model 8525) used for UFP collection.

The Condensation Particle Counter (CPC) operates based on photometric principle. The device counts particles that are extracted by a pump, through the use of a specific laser beam. In order for such smaller particles to be counted, they are subjected to an enlargement process, in which alcohol vapour condenses around them and leads to the formation of higher size droplets. This is done by passing the particles through a saturator tube which contains 100% reagent grade isopropyl alcohol in vapour form, which mixes with the particles. Afterwards, this mixture is led into a condenser, where the condensation into droplets occurs. Having proper dimensions for detection, these droplets pass through a focused laser beam, providing the counting of the particles by a photodetector. Considering the sampler working principle it is necessary to immerse repetitively its cylinder with isopropyl alcohol in order to prevent the sampler from malfunctioning and thus misleading particle number concentration measurements. Thus a measuring period of 8 h was used in all homes, as previously determined as a safe duration without the necessity of repetitive use isopropyl alcohol; UFP logging interval was 60s continuously.

The sampling apparatuses were typically placed in the corner of each room as far as possible from windows or doors, and other predictable sources of UFP. However, in some cases compromises had to be made, in order to maintain proper equipment functions and children safety. As for outdoor sources, the selected locations for the equipment were mainly terraces in a place as shield as possible so the equipment was protected from meteorological conditions such as the rain.

In order to better characterize the microenvironments, indoor temperature and relative humidity were continuously measured (with logging interval of 5 min) by TSI Q-Trak Indoor Air Quality Monitor, model 7575 (*Figure 5-b*). The real-time monitoring of some indoor gaseous pollutants such as NO_2 and O_3 was made by Series 500 – Portable Indoor Air Quality Monitor (*Figure 5-b*).

a) b)

Figure 5 - The equipment used for continuous sampling of: a) indoor pollutants (NO_2 and O_3); and b) physical parameters (temperature and relative humidity).

All used samplers and equipment were calibrated in accordance with the manufactures' guidelines and recommendations.

3.6 Statistical methods

A descriptive statistics analysis (mean, range, standard deviation) was performed to characterize the main parameters analyzed.

The Kolmogorov-Smirnov test was used to verify UFP data distribution for normality. Since a non-Gaussian distribution was observed, non-parametric tests were used to process the data. Inferential statistics between two variables were performed using the Mann-Whitney test, while for 3 or more variables the Kruskal-Wallis test was used. A significant difference was considered if $p<0.05$.

All statistical analyses were performed using a statistical analysis software package, SPSS (Statistical Package for the Social Sciences), version 20 (IBM).

3.7 References

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4. Results and discussions

4.1 Indoor Environments

Regarding indoor home environments, the overall levels of UFP ranged from 1.72×10^3 particles cm^{-3} registered in some of the bedrooms to the highest value of 3.62×10^5 particles cm^{-3} registered in the only kitchen analyzed. *Figure 6* presents the means of UFP number concentrations in both bedrooms and living rooms of the 14 homes.

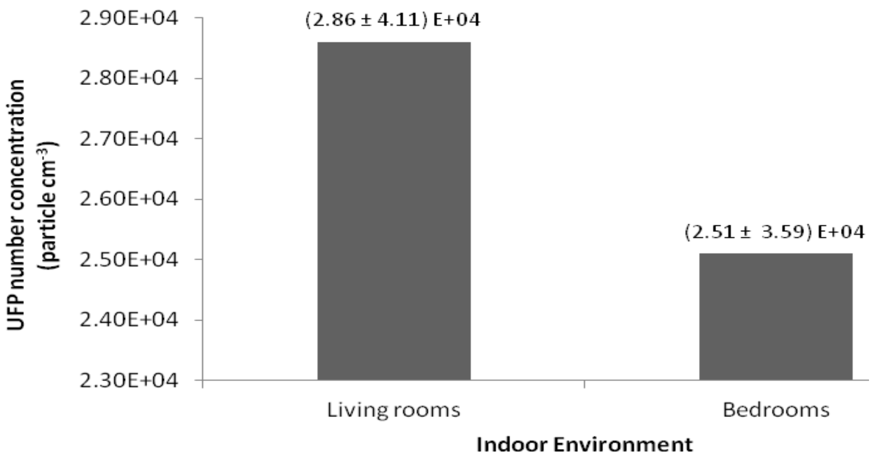

Figure 6 - Ultrafine particle number concentration means and standard deviations for the two indoor environments measured (particle cm^{-3}). (n=14 homes)

Ultrafine particle number concentrations for the indoor environments were statistically compared. Mann-Whitney test showed that there were statistically significant differences between the bedrooms and the living rooms.

Results of the average and range of indoor ultrafine particle number concentrations for each home are shown in *Table 5*. These results include the three indoor environments where the measurements were done, namely the bedrooms, the living rooms and one kitchen.

Table 5 - Summary of the mean of indoor particle number concentrations for each home (particles cm^{-3}).

Home	Bedroom (n=14)		Living room (n=10)		Kitchen (n=1)		Total Indoor (n=10)	
	Mean	Range (Min-Max)	Mean	Range (Min-Max)	Mean	Range (Min Max)	Mean	Range (Min-Max)
C07	5.54×10^3	$2.34 \times 10^3 – 2.92 \times 10^4$	1.12×10^4	$1.83 \times 10^3 – 1.29 \times 10^5$	-	-	8.39×10^3	$1.83 \times 10^3 – 1.29 \times 10^5$
C10	6.14×10^4	$1.10 \times 10^4 – 2.00 \times 10^5$	8.83×10^4	$2.48 \times 10^4 – 1.94 \times 10^5$	-	-	7.49×10^4	$1.10 \times 10^4 – 2.00 \times 10^5$
C11	4.49×10^3	$2.57 \times 10^3 – 7.17 \times 10^3$	5.69×10^3	$3.39 \times 10^3 – 9.59 \times 10^3$	-	-	5.09×10^3	$2.57 \times 10^3 – 9.59 \times 10^3$
C21	5.48×10^3	$3.06 \times 10^3 – 1.20 \times 10^4$	6.00×10^3	$2.85 \times 10^3 – 1.28 \times 10^4$	-	-	5.74×10^3	$2.85 \times 10^3 – 1.28 \times 10^4$
C25	8.44×10^4	$1.23 \times 10^4 – 2.65 \times 10^5$	6.55×10^4	$1.91 \times 10^4 – 1.47 \times 10^5$	9.68×10^4	1.75×10^4 3.62×10^5	8.28×10^4	$1.23 \times 10^4 – 3.62 \times 10^5$
C27	8.27×10^3	$5.40 \times 10^3 – 1.23 \times 10^4$	9.06×10^3	$8.04 \times 10^3 – 1.03 \times 10^4$	-	-	8.31×10^3	$5.40 \times 10^3 – 1.23 \times 10^4$
C34	1.95×10^4	$9.76 \times 10^3 – 4.03 \times 10^4$	2.11×10^4	$1.16 \times 10^4 – 1.03 \times 10^5$	-	-	2.03×10^4	$9.76 \times 10^3 – 1.03 \times 10^5$
C35	1.02×10^4	$4.82 \times 10^3 – 1.61 \times 10^4$	-	-	-	-	-	-
C41	1.41×10^4	$3.67 \times 10^3 – 3.06 \times 10^4$	2.81×10^4	$6.92 \times 10^3 – 1.27 \times 10^5$	-	-	2.11×10^4	$3.67 \times 10^3 – 1.27 \times 10^5$
C45	8.54×10^3	$2.72 \times 10^3 – 1.19 \times 10^5$	8.57×10^3	$3.57 \times 10^3 – 8.20 \times 10^4$	-	-	8.55×10^3	$2.72 \times 10^3 – 1.19 \times 10^5$
C48	4.68×10^3	$1.72 \times 10^3 – 1.10 \times 10^4$	2.39×10^4	$3.91 \times 10^3 – 1.00 \times 10^5$	-	-	1.40×10^4	$1.72 \times 10^3 – 1.00 \times 10^5$
C50	2.39×10^4	$3.91 \times 10^3 – 1.00 \times 10^5$	-	-	-	-	-	-
C53	6.62×10^3	$6.75 \times 10^3 – 2.03 \times 10^4$	-	-	-	-	-	-
C55	3.07×10^4	$3.69 \times 10^3 – 1.81 \times 10^5$	-	-	-	-	-	-
Total	**2.51×10^4**		**2.86×10^4**		-		**2.78×10^4**	

n = number of homes

The overall mean of UFP levels in bedrooms of all 14 homes was 2.51×10^4 particle cm^{-3}. In living rooms the overall mean of all sampled homes was 2.86×10^4 particle cm^{-3}. C25 and C10 were the homes that exhibited the highest UFP levels in both bedrooms and living rooms. In C10 indoor smoking was allowed in at least one division of the home and pets were present. In C25 the possible source of indoor UFP was probably due to pets.

The UFP measured in kitchen of home C25 shows higher mean concentration in comparison with the averages of other compartments of that. These results show contribution of combustion sources (i.e. cooking) on the presence of UFP indoors.

The lowest levels of UFP were identified at bedrooms of home C48 and living room of C11 probably due to a larger room area and bigger air volume and the presence of fewer room occupants.

4.1.1 Ultrafine Particle Number Concentrations: Comparison with International Studies

Table 6 provides comparison of indoor UFP levels with those from other studies.

Table 6 - Results of indoor UFP number concentration of other international studies.

Country	Mean	Study Conditions	Reference
Portugal	2.78×10^4 particles cm^{-3}		This study
Sweden	4.50×10^3 particle cm^{-3} (1) 2.40×10^3 particle cm^{-3} (2)	22 randomly selected homes; measurements conducted for 7 consecutive days;	(Isaxon, Gudmundsson et al. 2015)
Italy	8.64×10^3 particle cm^{-3}	one non-specified residential site; 24 h measurements; sampling periods: 1st campaign from 22 February 2012 to 7 March 2012; 2nd campaign from 16 to 30 April 2012; 3rd campaign from 28 May to 12 June;	(Zauli Sajani, Ricciardelli et al. 2015)
Singapore	8.5×10^3 particle cm^{-3}	1 home (living room, master bedroom, bedroom, and corridor); measurements from May 12 to May 23, 2004, with 2 days of sampling	(Balasubramanian and Lee 2007)
USA	4.59×10^3 particle cm^{-3} (3) 1.01×10^4 particle cm^{-3} (4) 6.05×10^3 particle cm^{-3} (5)	1 occupied suburban house; measurements between November 21, 1997 and December 31, 2000;	(Wallace 2006)
Canada	2.70×10^3 particle cm^{-3} (6) 3.73×10^3 particle cm^{-3}(7) 2.58×10^3 particle cm^{-3}(8)	45 non-smoking homes and 49 homes of asthmatic children; measurements of 10 mins every hour for 5 consecutive days	(Kearney, Wallace et al. 2011)

[1] During occupancy time
[2] During non-occupancy time
[3] 10 – 18 nm
[4] 18 - 50 nm
[5] 50 - 100 nm
[6] Summer 2005
[7] Winter 2006
[8] Summer 2006

The results in table 6 shows that indoor UFP measured in various European countries such as Sweden, Italy reported significantly lower levels of UFP than in present study. Similarly, studies conducted in Singapore, Canada and USA presented lower means of UFP. Seasonal influences, meteorological conditions, level of urbanization, and overall development of area where the homes were located could account for some of these differences (Morawska et al., 2009). Other study design (sampling period, duration, number of homes could also contribute to the obtained differences (Morawska et al., 2013). In addition, differences in the measured particle range, especially in terms of lower cut-off size could also account for some of the existent results (Kumar et al., 2010).

4.3 Indoor and Outdoor Concentrations

Outdoor mean number concentrations of ultrafine particles in the available homes are shown in *Table 7*. In order to analyze the influence of outdoor sources on indoor UFP levels, indoor/outdoor (I/O) ratios were calculated and are also presented in *Table 7*. These measurements allow a better understanding of indoor ultrafine particle concentration and particle behavior in terms of penetration from outdoor sources and transportation.

Table 7 - Summary of the average outdoor UFP number concentrations for each home (particles cm^{-3}) and I/O ratios.

Home **(n=8)**	**Outdoor**		I/O Concentration Ratio			
			Bedroom		**Living room**	
	Mean	Range (Min-Max)	Mean	Range (Min-Max)	Mean	Range (Min-Max)
C07	1.12×10^4	$1.83 \times 10^3 – 1.29 \times 10^5$	0.64	0.04 - 1.96	1.00	1.00 – 1.00
C10	2.66×10^4	$5.75 \times 10^3 – 7.35 \times 10^4$	3.81	0.33 - 26.86	5.00	0.83 - 21.88
C11	5.27×10^3	$2.41 \times 10^3 – 1.54 \times 10^4$	0.94	0.32 - 2.58	1.19	0.35 - 3.24
C21	7.97×10^3	$3.31 \times 10^3 – 1.58 \times 10^4$	0.75	0.28 - 2.64	0.83	0.28 - 2.62
C27	1.60×10^4	$6.52 \times 10^3 – 3.15 \times 10^4$	0.58	0.27 - 1.42	1.09	0.97 - 1.29
C41	7.04×10^3	$1.13 \times 10^3 – 2.01 \times 10^4$	2.38	0.50 - 7.78	4.38	1.04 - 20.94
C45	8.59×10^3	$3.39 \times 10^3 – 2.08 \times 10^4$	1.26	0.24 - 18.38	1.18	0.44 - 13.57
C55	2.21×10^4	$2.11 \times 10^3 – 5.54 \times 10^4$	2.89	0.11 - 22.53	-	-

n = number of homes

Home C10 shows the highest levels of outdoor UFP concentration. This home was located close to VCI, a road with high vehicle density. Therefore, the overall high levels of UFP levels culd be due to contribution from traffic emissions; previously it was shown that vehicle traffic is a significant source of UFP in Porto Metropolitan Area (Slezakova et al., 2014).

In order to further analyse the influence of outdoor concentrations in indoor UFP levels the indoor/outdoor ratios (I/O) were calculated.

- If I/O < 1 – Indoor concentration values were probably mostly affected by outdoor sources through transportation and infiltration mechanisms (i.e. outdoor air was the predominant source of UFP indoors).

- If I/O > 1 – Indoor concentration values were mostly due to indoor UFP producing activities (i.e. indoor sources were the predominant contributor to indoor UFP).

In homes C10, C41, and C55 (for bedrooms) the I/O ratios were much higher than 1, which suggest strong contribution of UFP mainly from indoor sources; additionally confined space inside the homes might not enable the dispersion of UFP. The highest I/O ratio for both bedroom and living room were found for home C10 where smoking was allowed. These results thus show the strong contribution of that source to indoor levels of UFP. Furthermore, although the respective parents did not smoke directly in child bedroom and were attentive of child well-being, these results clearly demonstrate that if smoking is allowed and conducted inside a home, its emissions easily penetrate to and influence all indoor microenvironments.

A I/O ratio value close to 1 leads to the conclusion that overall indoor and outdoor concentrations were similar. As seen in Table 7 I/O ratio of UFP measured bedrooms of majority of the homes, namely C07, C11, C21, C27 and C45 were below 1 (or close to unity for C45) which means that the activities conducted indoors of the respective microenvironments had a significantly lower effect on indoor UFP levels. These results are somewhat assuring considering the long period of time that children spent in their bedrooms. In contrast, the I/O ratios of UFP obtained for the living rooms tend to be much higher probably due to the number of occupants; majority of family joined in the livings rooms and conducted there most of their daily family life (watching television, studying, socializing, etc.). Furthermore, living room often included dining area and during weekends when the UFP sampling was conducted they were often used to serve the main meals (lunches and dinner).

Figure 7 shows an example of time series of the indoor and outdoor particle concentrations and their ratios for a randomly selected home. It can be seen that there was a positive correlation between indoor and outdoor UFP concentrations, indicating that indoor concentrations were influenced by outdoor sources under normal ventilation conditions. The period between 11:02 and 11:31 shows much higher values of indoor UFP nmber concentration which indicates a strong contribution of UFP from indoor sources.

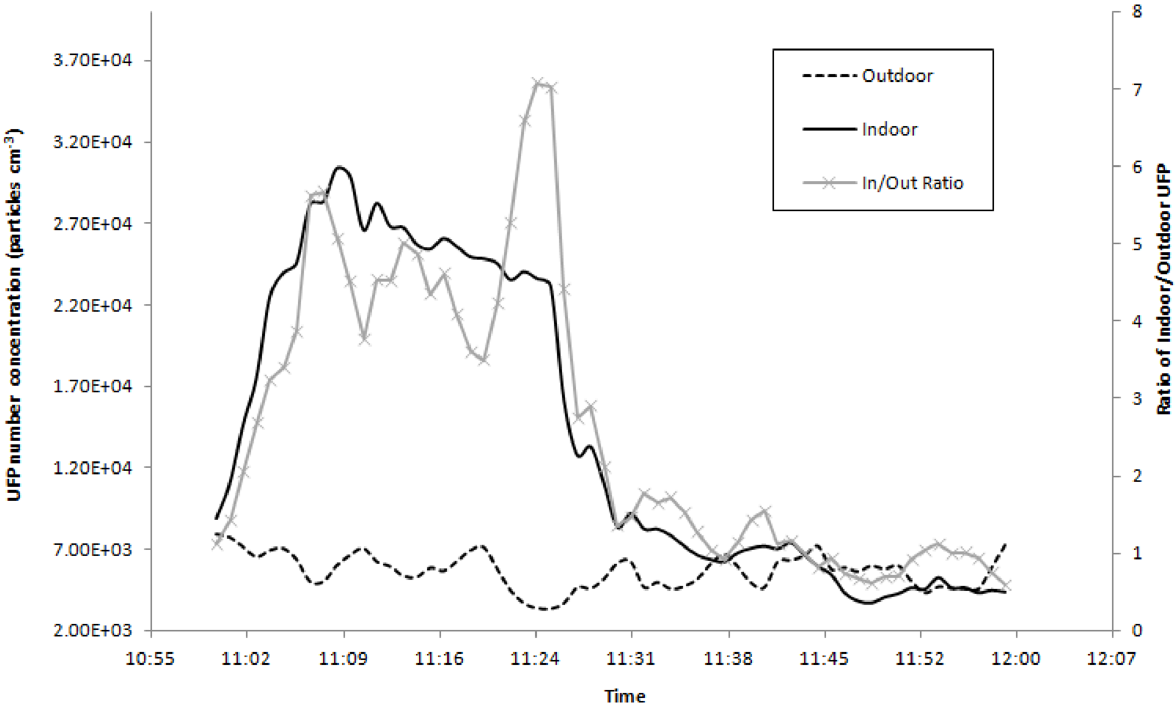

Figure 7 - Time series of the indoor and outdoor particle concentrations as well as the variation of indoor to outdoor concentration ratios.

4.4 References

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5. Conclusions and Future Perspectives

5.1 Conclusions

The execution of this study had led to the following conclusions.

Statistically significant differences between the levels of ultrafine particles levels measured in the bedrooms and in the living rooms of 14 Portuguese homes were obtained.

The indoor UFP concentrations in residences are fully dependent on the existent sources and living habits of the occupants. While there are different sources responsible for the origin of UFP, two main sources were identified in studied homes related with combustion activities, namely smoking and cooking. Nonetheless, the presence of pets and as well as room characteristics (volume, openable windows, etc.) had also a significant influence on indoor UFP levels. The higher number concentrations of UFP were found in indoor spaces with smaller air volume.

The information on UFP in indoor home environments is rather limited. Comparison with other international studies showed higher UFP levels in Portuguese homes than those in other studies conducted in Europe, or in Singapore and Canada. One study from USA reported higher levels UFP; however, study designs varied significantly among the work reported in literature.

Regarding outdoor UFP, the highest levels were found for homes situated in areas with high vehicle density in streets and roads surrounding these homes, thus showing that traffic emissions highly contribute to outdoor ultrafine particle levels. Indoor to outdoor ratios of UFP indicated that in bedrooms (where children spend most of their time) the activities conducted had a significantly lower effect on indoor UFP levels than in living rooms where indoor sources were the major contributors for indoor UFP levels, possibly due to family activities that were often conducted here, especially on weekends .

5.2 Future Perspectives

UFP can originate from many different indoor sources and activities of occupants. Therefore, future studies should focus on assessment of the identification of potential indoor UFP sources using the specific information of the common practices and activities held at home (preferably via questionnaires).

In order to improve knowledge about indoor UFP, future work should include measurements of other indoor parameters such a temperature, relative humidity, and concentrations of relevant indoor pollutants (VOCs, CO_2 and CO) should be considered. Also a higher number of homes would benefit the results obtained.

It would also be important to consider a wider range of measurements periods. The repetition of the sampling campaign during different seasons would be also useful, since UPF number concentrations outdoors, and consequently indoors, vary significantly between seasons.

APPENDIX

A.Questionnaires

In order to better understand the conditions of the sampling campaign and the possible factors influencing the study, two questionnaires were filled.

A.1 Parents’ diary

This diary, presented in this section, was given out to the parents or a responsible adult, who had to register the time (and duration) of specific activities/sources that were conducted/used in the evaluated compartments of the house.

Gostaríamos que preenchesse a seguinte checklist com informação relacionada com a casa e o ambiente interior. Por favor assinale com um "X" as opções que se apliquem referentes a cada um dos dias da semana.

Sábado

		Sábado																							
	Horas	**1**	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**	**10**	**11**	**12**	**13**	**14**	**15**	**16**	**17**	**18**	**19**	**20**	**21**	**22**	**23**	**24**
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Outros ______________________________																									

Checklist code_pais/encarregados de educação

Domingo

		Domingo																							
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Checklist code_pais/encarregados de educação

2.º Feira

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3.º Feira

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Checklist code_pais/encarregados de educação

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Checklist code_pais/encarregados de educação

Informações adicionais:

(por exemplo: outro tipo de limpeza, remodelação, decoração, presença de animais de estimação, atividades desenvolvidas, …)

Checklist code_pais/encarregados de educação

A.2 House Characteristics Checklist

The house checklist dedicated to external and internal building description and filled by a researcher of the study project is presented following.

PROJECTO

ARIA

CHECKLIST **DA HABITAÇÃO**

(A PREENCHER POR TÉCNICOS RESPONSÁVEIS)

PT	E			S	-			

PT	C			

Informação geral do edifício

Identificação

Vivenda	☐
Vivenda geminada	☐
Apartamento	☐
Outro (especifique)________________	☐

Morada:

____

____

Coordenadas GPS: ____

Contacto:

Nome:____

Telefone:___________________________ e-mail: ___________________________________

Orientação do edifício, sombreamento do edifício por outros edifícios na proximidade: (esboço do edifício e envolvente, fotografia, imagem do google, …)

Responsável______________________________________Data________________

A. Edifício

1. Caracterização da envolvente

1.1. [obs.] Localização do edifício

Urbana	Zona industrial	
	Zona industrial/área residencial	
	Zona comercial	
	Zona comercial/área residencial	
	Centro da cidade, densamente ocupada com habitações	
	Zona urbana com ou sem jardins	
Zona suburbana com jardins		
Rural	Vila	
	Área pouco urbanizada (baixa densidade de habitações)	

1.2. [obs.] Fontes de poluição próximas

Nenhuma	
Parques de estacionamento	
Garagens	
Acesso direto ao parque de estacionamento	
Proximidade de vias com tráfego intenso	
Proximidade de autoestradas	
Proximidade a postos de gasolina	
Proximidade de indústrias (até 1 Km)	
Proximidade de torres de arrefecimento	
Construção sobre aterro sanitário	
Proximidade de local de gestão de resíduos (até 1 Km)	
Proximidade de campos agrícolas (até 1 Km)	
Outro (especifique) ______________________________	

2. Caracterização do edifício

2.1. [obs.] Número de pisos (Total__________)

Acima do solo	
Abaixo do solo	

2.2. [obs.] Material de construção do edifício

Tijolo	
Outro (especifique)	

2.3. [ask] Existe para o edifício algum certificado*?

Não	
Sim	
Qual ____________________	

* Legislação, Regulamento (Performance energética, QAI, sustentabilidade, …)

3. Potenciais fontes de poluentes no interior

3.1. [obs.] Distância entre o edifício e o contentor de lixo no exterior (aproximadamente)________________m

3.2. [obs.] Utilização de espaços (no caso de edifícios multifamiliares - apartamentos)

Farmácia	
Padaria/pastelaria	
Restaurante	
Papelaria	
Serviços	
Comércio	
Outro (especifique)	

3.3. Utilização de pesticidas (desde o nascimento da criança)

		Interior		Exterior	
		Frequência	Última data	Frequência	Última data
Não		--	--	--	--
Sim, no passado			/ /		/ /
Sim, no presente			/ /		/ /
Razão para ter deixado de utilizar	____________________ ____________________ ____________________				

B. Habitação

1. Caracterização da habitação

1.1. [ask] Há quantos anos vive nesta habitação: _________________

1.2. [obs.] Número do andar: ______________________________

1.3. [obs.] Esboço da habitação e identificação dos diversos compartimentos (cozinha, sala, WC, quartos, …)

Esboço do layout da habitação	Compartimentos	Nr.	Área [m^2]	Janelas	
				Nº	Área [m^2]
	Cozinha (cz)				
	Casa de banho completa (cb)				
	WC (WC)				
	Corredor (c)				
	Quarto (q)				
	Sala de estar (s)				
	Lavandaria (l)				
	Despensa (d)				

1.4. [ask] Número total de ocupantes a viver permanentemente na habitação (especificando o grupo etário):

Número total de ocupantes	
Número total de ocupantes por faixa etária	
< 6 anos	
6-12 anos	
13-17 anos	
18-30 anos	
31-64 anos	
>65 anos	

1.5. [ask] É permitido fumar no interior da habitação

		Identificação dos fumadores		
Não		Pai	Mãe	Outro
Sim				

1.6. [ask] Local onde é permitido fumar

		N.º de fumadores	N.º de cigarros/por dia	Período do dia
Cozinha				
WC				
Corredor				
Quarto				
Sala de estar				
Lavandaria				
Despensa				
Varanda				
Outro local				

1.7. [ask] Os materiais utilizados no interior da habitação são materiais de baixa emissão? (classificação por sistema de rotulagem reconhecida, identificação: GEV, AgBB, …)

Adesivos e colas	
Tintas e revestimentos	
Materiais para tetos e paredes	
Materiais para o pavimento	
Compósitos de madeira e produtos de fibras agrícolas	
Mobiliário	
Outro (especifique)	

1.8. [obs.] Presença visível de rachaduras na estrutura do edifício?

Não	
Sim	
Se sim, onde? ____	

1.9. [ask] Foi feita alguma modificação no interior da habitação desde o nascimento da criança?

	Data	Identificação do compartimento	Motivo
Pavimento	____/___/____		
Isolamento	____/___/____		
Paredes	____/___/____		
Teto/telhado	____/___/____		
Sistema de aquecimento	____/___/____		
Instalação e/ou remoção de equipamento de AC	____/___/____		
Sistema de ventilação	____/___/____		
Instalação e/ou remoção de equipamento de ventilação mecânica	____/___/____		
Janelas	____/___/____		
Mobiliário	____/___/____		
Outro (especifique) ________________	____/___/____		

C. Local de monitorização

Quarto	
Outro (especifique)	____________________

2. Caracterização do ambiente exterior

2.1. [obs.] A criança vive na proximidade de vias com tráfego intenso (200m ou perto__________m (especificar)?

Não	
Sim	
Autoestrada	
Estrada	
Avenida	
Outro (especifique)	

3. Caracterização do local de monitorização

3.1. [ask] Identificação do local onde a criança dorme

Quarto	
Quarto dos pais	
Sala de estar	
Outro (especifique)	

3.2. [obs.] Área do espaço______________________m^2

3.3. [obs.] Pé direito____________________________m

3.4. Número total de ocupantes permanentes no local de monitorização: ________________________

3.5. Janelas e área envidraçada

Janelas			Área envidraçada	
Número	Orientação*	Área (m^2)	%	Orientação*

3.6. [obs.] Caixilharia das janelas

Metal	
Madeira	
PVC	
Alumínio	
Outro (especifique)	

3.7. [obs.] Tipo de envidraçados

Vidro simples		
Vidro duplo	Com preenchimento de árgon ou outro	
	Com revestimento	
	Com painel interno matizado	
Vidro triplo		
Outro (especifique)		

3.8. [obs.] Dispositivos de proteção solar (estores)

Nenhum	
Apenas no lado sul	
Outras fachadas	
Exterior	
Interior	

3.9. [obs.] Os dispositivos de proteção solar dificultam a utilização das janelas ou diminuem a capacidade de ventilação do local?

Não	
Sim	

3.10. [obs.] Identificação do revestimento do teto e paredes

	Teto	Paredes
Cimento		
Tinta		
Papel de parede		
Material sintético		
Ladrilhos de fibra mineral		
Ladrilhos de fibra de madeira/cortiça		
Madeira		
Gesso		
Outro (especifique)		

3.11. [obs.] Identificação do revestimento do pavimento

Cimento	
Carpete	
Material sintético (linóleo, vinil, ...)	
Parquet	
Pedra/azulejo	
Madeira/cortiça	
Outro (especifique)	

3.12. [ask] Abertura de portas (número) e janelas (número e orientação) (habitualmente)

	Portas			Janelas		
	Abertas	Parcialmente aberta	Fechadas	Abertas	Parcialmente aberta	Fechadas
Início da manhã						
Durante a manhã						
À tarde						
À noite						
Durante o período de sono						

* N (norte), S (sul), E (este), W (oeste)

3.13. [obs.] Identificação do material do mobiliário

Madeira	
Revestimento fino de madeira	
Madeira compensada	
Têxtil	
Metal	
Plástico	
Outro (especifique)	

4. Patologias relacionadas com o aparecimento de fungos e/ou bolores

4.1. [obs., ask] Nos últimos 12 meses ocorreram problemas de fugas de água e/ou inundações?

	Local monitorizado	Outro compartimento
Não		
Sim		
Telhado	/ /	/ /
Janelas	/ /	/ /
Fachada	/ /	/ /
Cave	/ /	/ /
Outro (especifique) _	/ /	/ /

4.2. [obs.] Danos causados por incêndio

	Local monitorizado	Outro compartimento
Não		
Sim	/ /	/ /
Extensão dos danos		
Em todo o edifício		
Em áreas limitadas		
Danos nos outros pisos		

4.3. [obs., ask] Crescimento visível de bolores

	Local monitorizado	Outro compartimento
Não		
Sim, no passado		
Local (paredes, teto, pavimento)		
Extensão		
Sim, à data da avaliação		
Local (paredes, teto, pavimento)		
Extensão		

4.4. [obs.] Outros problemas de humidade/bolores

	Local monitorizado		Outro compartimento	
	Não	Sim	Não	Sim
Odor a mofo				
Manchas visíveis de humidade nas paredes, teto e/ou pavimento				
Manchas ou descoloração amarela no pavimento de plástico				
Manchas pretas no pavimento				

4.5. [obs., ask] Tendência para a formação de condensação nas janelas/envidraçados

	Local monitorizado	Outro compartimento
Não		
Sim		
No interior (vidros)		
Na caixilharia		

5. Aquecimento

5.1. [obs.] Existe sistema de aquecimento?

	Local monitorizado	Outro compartimento
Não		
Sim		
Se sim, em funcionamento?		

5.2. [obs.] Sistema de aquecimento

	Local monitorizado	Outro compartimento
Apenas aquecimento		
Aquecimento + aquecimento de águas sanitárias		

5.3. [obs.] Identificação do tipo de sistema de aquecimento

		Local monitorizado	Outro compartimento
Radiadores de água quente ou convectores			
Radiadores eléctricos ou convectores			
Piso radiante			
Lareiras	Aberto		
	Fechado		
Outro (especifique) _			

5.4. [obs.] Os aquecedores estão localizados de baixo das janelas?

	Local monitorizado	Outro compartimento
Não		
Sim		

6. Ventilação

6.1. [obs.] Estratégia de ventilação

	Local monitorizad	Outro compartimento
Natural		
Natural assistida (exaustão)		
Mecânica		

6.2. [obs.] As janelas podem ser abertas?

	Local monitorizado	Outro compartimento
Não		
Sim		
Todas (% de abertura)		
Algumas (número e % de abertura)		
Os ocupantes não estão autorizados a abrir as janelas		

7. Fontes de poluentes no interior

7.1. [obs.] Identificação do número de equipamentos elétricos

	Local monitorizado		Outro compartimento	
	Existe	Em utilização	Existe	Em utilização
Rádio				
Computador				
Impressora				
Fotocopiadora				
Televisão				
Outro (especifique)				

7.2. [obs., ask] Identificação do número de:

	Local monitorizado		Outro compartimento	
	Existe	Em utilização	Existe	Em utilização
Purificador de ar (especifique)				
Humidificador				
Desumidificador				
Sprays				
Permanente (passivo ou elétrico)				
Ocasionalmente (pulverização)				
Frequência de utilização				

8. Limpeza

8.1. [ask] Plano de limpeza

Início da manhã	
Durante a manhã	
À tarde	
À noite	
Outro (especifique)	

8.2. [ask] As janelas são abertas durante a limpeza?

	Local monitorizado	Outro compartimento
Não		
Sim		

8.3. [ask] Frequência da limpeza e produtos utilizados

	Local monitorizado				Outro compartimento			
	Spray	Liquido	ID	Freq*	Spray	Liquido	ID	Freq*
Pavimento								
Lixivia ou detergente c/ lixivia								
Detergente s/ lixivia								
Encerar								
Varrer								
Aspirar Aspirador água								
Aspirador de saco								
Outro								
Paredes								
Lixivia ou detergente c/ lixivia								
Detergente s/ lixivia								
Outro								
Janelas/envidraçados								
Detergente c/ amoníaco								
Detergente s/ amoníaco								
Outro								
Mobiliário								
Com detergente								
Apenas pano (seco)								
Outro								

	Local monitorizado	Freq*	Outro compartimento	Freq*
Cestos do lixo esvaziados				
Lavar as cortinas				
Comentários adicionais				

*Frequência: a) diariamente; b) duas vezes por semana; c) uma vez por semana; d) uma vez por mês; e) uma vez por ano; f) nunca

9. Fontes de alergénios e sensibilização

9.1. [ask] Reformulações/alterações na habitação desde o nascimento da criança

	Especificar	Idade da criança	Compartimento	Razão
Colocar/remover plantas				
Ter/deixar de ter animais				
Colocar/remover carpetes				

9.2. [ask] Local onde dorme a criança

Cama com colchão	
Colchão no chão	
Sofá	
Sofá-cama	
Berço (sem colchão)	
Colchão baixo	
Outro (especifique)	

9.3. [ask] Tipo de cobertores, colcha e capas para almofadas

			Impermeáveis aos alergénios (S/N*, especificar a marca)
Cobertores	Algodão		
	Lã		
	Sintético		
	Outro		
Colcha	Algodão		
	Lã		
	Sintético		
	Outro		
Capas para almofadas	Algodão		
	Lã		
	Sintético		
	Outro		

* S (sim), N (não);

9.4. [obs., ask] Presença de almofadas

	Local monitorizado	Outro compartimento
Não		
Não, mas já existiram		
Há quanto tempo foram removidas		
Razão para terem sido removidas	____________________	____________________
Sim		
Número		
Material		
Penas		
Poliéster		
Espuma		
Têxtil natural		
Têxtil sintético		
Não sabe		
Outro (especifique)	____________________	____________________
Frequência de limpeza	____________________	____________________

9.5. [obs., ask] Presença de tapetes

	Local monitorizado	Outro compartimento
Não		
Não, mas já existiram		
Há quanto tempo foram removidos		
Razão para terem sido removidos	____________________	____________________
Sim		
Número		
Área		
Material		
Têxtil natural		
Têxtil sintético		
Outro (especifique)	____________________	____________________
Frequência de limpeza	____________________	____________________

9.6. [obs., ask] Presença de carpetes

	Local monitorizado	Outro compartimento
Não		
Não, mas já existiram		
Há quanto tempo foram removidas		
Razão para terem sido removidas	____________________	____________________
Sim		
Número		
Área		
Material		
Têxtil natural		
Têxtil sintético		
Outro (especifique)	____________________	____________________
Frequência de limpeza	____________________	____________________

9.7. [obs.] Presença de cortinas

	Local monitorizado	Outro compartimento
Não		
Não, mas já existiram		
Há quanto tempo foram removidas		
Razão para terem sido removidas	____________________	____________________
Sim		
Número		
Área		
Material		
Têxtil natural		
Têxtil sintético		
Outro (especifique)	____________________	____________________
Frequência de limpeza	____________________	____________________

9.8. [obs., ask] Brinquedos com pêlo

Não	
Não, mas já existiram	
Razão para terem sido removidos ______	
Sim	
Número	
Localização (prateleiras, espaço fechado, espalhados pelo espaço, …) ______	

9.9. [obs.] É visível a existência de pó acumulado no local monitorizado?

Não	
Sim	

9.10. [ask] Presença de animais de estimação na habitação à data da avaliação e no passado (após o nascimento da criança)

		N.º	Há quanto tempo/ durante quanto tempo	Interior		Exterior		Contacto com a criança (S/N)ǂ
				Dia	Noite	Dia	Noite	
Não		--	--	--	--	--	--	--
Não, mas já existiram*								
Sim								
Cão								
Gato								
Outro animal de pêlo								
Peixe/tartaruga								
Pássaro								
Roedores								
Outro (especifique)								
Razão para deixar de ter animais de estimação								

* Preencher as colunas relativas ao Sim; ǂ S (sim), N (não);

9.11. [ask] Presença de plantas à data da avaliação e no passado na habitação (após o nascimento da criança)

		N.º	Há quanto tempo/ durante quanto tempo	Interior	Exterior	Contacto com a criança (S/N)ǂ
Não		--	--	--	--	--
Não, mas já existiram*						
Sim						
Parietaria spp						
Oliveira						
Bétula						
Relva/erva (perguntar por cevada, centeio, aveia, milho, trigo)						
Outro (especifique)						
Razão para deixar de ter plantas						

* Preencher as colunas relativas ao Sim; ǂ S (sim), N (não);

D. Atividades da criança

1.1. [ask] Excluindo as atividades escolares quais as atividades da criança e o tempo despendido em cada uma delas (fora da habitação).

	Dias da semana	Fim-de semana	Tempo despendido (h/dia)

<u>Durante a semana:</u>

1.2. [ask] Em média, quantas horas a criança passa no interior da habitação?

	Tempo despendido (h/dia)
Segunda-feira	
Terça-feira	
Quarta-feira	
Quinta-feira	
Sexta-feira	

1.3. [ask] Em que compartimento da habitação a criança passa a maioria do tempo?

	Tempo despendido (h/dia)
Quarto	
Quarto da família	
Sala de estar	
Cozinha	
Escritório	
Outro (especifique)	

1.4. [ask] Quais as principais atividades da criança quando permanece na habitação (interior e exterior)?

	Tempo despendido (h/dia)	Compartimento da habitação/exterior
Jogar (computador, ps, …)		
Ver televisão		
Brincar		
Outro (especifique)		

Durante o fim-de-semana:

1.5. [ask] Em média, quantas horas a criança passa no interior da habitação?

	Tempo despendido (h/dia)
Sábado	
Domingo	

1.6. [ask] Em que compartimento da habitação a criança passa a maioria do tempo?

	Tempo despendido (h/dia)
Quarto	
Quarto da família	
Sala de estar	
Cozinha	
Escritório	
Outro (especifique)	

1.7. [ask] Quais as principais atividades da criança quando permanece na habitação (interior e exterior)?

	Tempo despendido (h/dia)	Compartimento da habitação/exterior
Jogar (computador, psp, …)		
Ver televisão		
Brincar		
Outro (especifique)		