Universidade de Aveiro Departamento de Ambiente e Ordenamento
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DANIELA SOFIA OLIVEIRA DIAS

MODELAÇÃO DA EXPOSIÇÃO A POLUENTES TÓXICOS RELACIONADOS COM O TRÁFEGO

EXPOSURE MODELLING TO TRAFFIC-RELATED AIR TOXIC POLLUTANTS

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Tese apresentada à Universidade de Aveiro para cumprimento dos requisitos necessários à obtenção do grau de Doutor em Ciências e Engenharia do Ambiente, realizada sob a orientação científica da Doutora Oxana Tchepel, Investigadora Auxiliar do Centro de Estudos do Ambiente e do Mar, Departamento de Ambiente e Ordenamento, da Universidade de Aveiro.

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palavras-chave

Exposição humana, poluição atmosférica, áreas urbanas, tráfego rodoviário, GPS, GIS, padrões de atividade-tempo.

resumo

Atualmente, a poluição atmosférica constitui uma das principais causas ambientais de mortalidade. Cerca de 30% da população europeia residente em áreas urbanas encontra-se exposta a níveis de poluição atmosférica superiores aos valores- limite de qualidade do ar legislados para proteção da saúde humana, representando o tráfego rodoviário uma das principais fontes de poluição urbana. Além dos poluentes tradicionais avaliados em áreas urbanas, os poluentes classificados como perigosos para a saúde (Hazard Air Pollutants - HAPs) têm particular relevância devido aos seus conhecidos efeitos tóxicos e cancerígenos. Neste sentido, a avaliação da exposição torna-se primordial para a determinação da relação entre a poluição atmosférica urbana e efeitos na saúde.

O presente estudo tem como principal objetivo o desenvolvimento e implementação de uma metodologia para avaliação da exposição individual à poluição atmosférica urbana relacionada com o tráfego rodoviário. De modo a atingir este objetivo, foram identificados os parâmetros relevantes para a quantificação de exposição e analisados os atuais e futuros potenciais impactos na saúde associados com a exposição à poluição urbana. Neste âmbito, o modelo ExPOSITION (EXPOSure model to traffIc-relaTed aIr pOllutioN) foi desenvolvido baseado numa abordagem inovadora que envolve a análise da trajetória dos indivíduos recolhidas por telemóveis com tecnologia GPS e processadas através da abordagem de *data mining* e análise geo-espacial. O modelo ExPOSITION considera também uma abordagem probabilística para caracterizar a variabilidade dos parâmetros microambientais e a sua contribuição para exposição individual. Adicionalmente, de forma a atingir os objetivos do estudo foi desenvolvido um novo módulo de cálculo de emissões de HAPs provenientes do transporte rodoviário.

Neste estudo, um sistema de modelação, incluindo os modelos de transporte-emissões-dispersão-exposição, foi aplicado na área urbana de Leiria para quantificação de exposição individual a PM2.5 e benzeno. Os resultados de modelação foram validados com base em medições obtidas por monitorização pessoal e monitorização biológica verificando-se uma boa concordância entre os resultados do modelo e dados de medições. A metodologia desenvolvida e implementada no âmbito deste trabalho permite analisar e estimar a magnitude, frequência e inter e intra-variabilidade dos níveis de exposição individual, bem como a contribuição de diferentes microambientes, considerando claramente a sequência de eventos de exposição e relação fonte-recetor, que é fundamental para avaliação dos efeitos na saúde e estudos epidemiológicos. O presente trabalho contribui para uma melhor compreensão da exposição individual em áreas urbanas, proporcionando novas perspetivas sobre a exposição individual, essenciais na seleção de estratégias de redução da exposição à poluição atmosférica urbana, e consequentes efeitos na saúde.

keywords

Human exposure, air pollution, urban areas, road traffic, modelling, GPS, GIS.

abstract

Currently, air pollution represents one of the main environmental causes of mortality. About 30% of European citizens in urban areas are exposed to air pollution levels that exceed the air quality limits set by the legislation for the protection of human health, with road transport being the most significant pollution source. In addition to the traditional air pollutants evaluated in urban areas, the hazardous air pollutants (HAPs) has been the subject of particular concern because of their known toxic and carcinogenic effects. In this sense, the evaluation of exposure becomes essential in determining the relationship between urban air pollution and health effects.

The main objective of the current study is the development and implementation of a consistent approach for the quantification of individual exposure to traffic-related air pollutants. For this purpose, relevant parameters of exposure quantification were identified and the current and future potential impacts on human health associated with exposure to urban air pollution were analysed. In this context, the ExPOSITION model (EXPOSure model to traffIc-relaTed aIr) was developed by using a novel approach based on the trajectory analysis of the individuals collected by mobile phones with GPS and processed using the data mining approach and geo-spatial analysis within GIS. Also, the ExPOSITION model considers a probabilistic approach to characterize the variability of microenvironmental parameters and its contribution to personal exposure. Additionally, in order to achieve the objectives of the current study, a new module to quantify emissions of traffic-related HAPs was developed.

In this study, a modelling system, including transport-emissions-dispersion-exposure models was applied to the Leiria urban area for quantification of individual exposure to PM2.5 and benzene. The modelling results were validated based on measurements obtained by personal monitoring and biological monitoring evidencing a good agreement between the model results and measurement data. The methodology developed and implemented in this work allows to estimate and analyse the magnitude, frequency and the inter and intra-variability of personal exposure levels, as well as the contribution of different microenvironments, clearly addressing the sequence of exposure events and source-receptor relationship, which is essential for health impact assessment and epidemiological studies. This research work contributes to a better understanding of individual exposure in urban areas, providing new perspectives on individual exposure, essential in the selection of strategies to reduce exposure to urban air pollution and related health effects.

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LIST OF ORIGINAL PUBLICATIONS

This thesis is based on the work contained in following original publications:

I. Tchepel O., Dias D. (2011) Quantification of health benefits related with reduction of atmospheric PM10 levels: implementation of population mobility approach. International Journal of Environmental Health Research. 21, 189-200. doi: 10.1100/2012/409546

II. Dias D., Tchepel O., Carvalho A., Miranda A.I., Borrego C. (2012) Particulate matter and health risk under a changing climate: assessment for Portugal. Scientific World Journal. Volume 2012, Article ID 409546, 10 pages. doi: 10.1100/2012/409546

III. Tchepel O., Dias D., Ferreira J., Tavares R., Miranda A.I., Borrego C. (2012) Emission modelling of hazardous air pollutants from road transport at urban scale. Transport. 27, 299-306. doi:10.3846/16484142.2012.720277

IV. Dias D., Tchepel O. (submitted to publication) Modelling of human exposure to air pollution in the urban environment: A GPS based approach. Environmental Science and Pollution Research. Manuscritpt Nº ESPR-D-13-00249

V. Tchepel O., Dias D., Costa C., Santos B.F., Teixeira J.P. (submitted to publication) Modelling of human exposure to benzene in urban environments. Atmospheric Environment. Manuscritpt Nº ATMENV-D-13-00451R1

ABBREVIATIONS

AD – Attributable Deaths
AMP – Porto Metropolitan Area
APHEIS – Air Pollution and Health: A European Information System
CO – Carbon Monoxide
CO_2 – Carbon Dioxide
CR – Concentration-Response functions
DGS – Direção Geral de Saúde
EEA – European Environment Agency
GHG – Greenhouse Gases;
GIS – Geographic Information System
GPS – Global Positioning System
HAPs – Hazardous Air Pollutants
HIA – Health Impact Assessment
INE – Instituto Nacional de Estatística
IPCC – International Panel on Climate Change;
IPCS – International Programme on Chemical Safety
LUR – Land-use regression models
NMVOC – Non-methane volatile organic compounds
NOx – Nitrogen Oxides
NO_2 – Nitrogen Dioxide
NRC – National Research Council
OECD – Organisation for Economic Co-operation and Development
pkm – passenger-kilometres
PM10 – Particulate matter with an equivalent aerodynamic diameter of less than 10 μm
PM2.5 – Particulate matter with an equivalent aerodynamic diameter of less than 2.5 μm
ppm – parts per million
RR – Relative Risk
TAD – Time-activity diary
t,t-MA – Trans, trans muconic acid
USEPA – U.S. Environmental Protection Agency
VOC – Volatile Organic Compound
WHO – World Health Organization

CHAPTER ONE

1. GENERAL INTRODUCTION

"The quality of the exposure data is still regarded as the Achilles' heel of air quality epidemiology – an improved understanding of personal exposure to air pollution is required."

Frank Kelly
Air Quality and Emissions conference,
Telford, 2013

Urban air pollution has emerged as one of the major and complex health problems and environmental concerns in Europe, with direct consequences for the health and well-being of European citizens. Considerable progress has been made in the past twenty years in improving urban air quality, but issues remain. Although, emissions of many air pollutants have decreased resulting for some pollutants in improved air quality, the European Environment Agency evaluated that about 30% of Europe's urban population is still exposed to air pollution concentrations exceeding the European Union (EU) air-quality limits set to protect human health (EEA, 2012a). By 2050, according to the Organisation for Economic Co-operation and Development (OECD), air pollution is anticipated to become the biggest environmental cause of mortality worldwide, overtaking the lack of clean water and poor sanitation (OECD, 2012). In this context, the World Health Organization considered urban air pollution as one of the most important global health priorities (WHO, 2011).

Road transport is likely to be the largest source of air pollutants that have a substantial impact on health (HEI, 2010). In addition to the traditional major air pollutants, such as carbon dioxide (CO_2), carbon monoxide (CO), nitrogen oxides (NOx) and non-methane volatile organic compounds (NMVOC), road transport is still one of the major sources of substances known as hazardous air pollutants (HAPs) especially in urban areas, contributing about 68% of HAPs total emissions (Tam and Neumann, 2004; HEI, 2010).

Given the need for understanding the impact of air pollutants on human health, outdoor air pollution measurements are performed. For this purpose, centrally located air quality monitoring stations are usually used to characterize air quality and considered as an indicator of human exposure to traffic-related air pollutant in urban areas, as needed for health impact assessment and for the design of air pollution control policies. However, individual exposure assessment based on fixed-site air measurements is unavoidably affected by assumptions implicit in the application of this approach. The challenge, however, is that exposure levels depend not only on environmental conditions, such as air pollution but also on the behaviour of an individual, making a personal exposure to urban air pollution a unique situation, occurring both in indoor and outdoor environments and thus is not straightforward to quantify.

Understanding of the complex chain of events, from traffic activities to emissions, ambient air quality, exposure and health effects would help decision-makers to focus their efforts and enable a more forceful reduction of adverse effects. Thus, the implementation of improved and comprehensive approaches to address exposure at the spatial and temporal scale imposed by the individual is required and has been identified as a priority area in the exposure research (Briggs, 2008; Nuckols et al., 2010; de Nazelle et al., 2011).

1.1. Human exposure in urban areas: origin and concepts

Urban areas with their complex pollution problems are identified as the main target of the current research. Human exposure to air pollution in urban areas and subsequent health effects results from a dynamic process and multifaceted iterations between the individual and urban air. In the following sections, crucial questions such as "*What are the main sources of air pollution and current air pollution levels in urban areas?*"; *"How human exposure to air pollution may be defined?"* and *"Why personal exposure assessment is needed?"* will be addressed. Also, important exposure-related concepts and key elements required to understand the human exposure science are described.

1.1.1. What are the main sources and current air pollution levels in urban areas?

Rapid urbanization and industrialization, increase in the road traffic and energy consumption, have contributed towards the increase in ambient air pollution concentrations and consequent deterioration of ambient air quality. Urban environment, where currently around 75% of the European population lives and this is projected to increase to about 80% by 2020 (EEA, 2010), is particularly affected. Air pollution levels are still rising on many fronts. However, air pollution is enacted on all geographical and temporal scales, ranging from strictly “here and now” problems related to human health, over regional phenomena with a time horizon of decades, to global phenomena, which over the next centuries can change the conditions for human being and environment over the entire globe. Although most environmental and health issues are not exclusive to urban areas, some are exacerbated within them, because of the specific urban complexity of interrelations between environmental, social and economic demands (RCEP, 2007; DEFRA, 2008). People in urban areas use more energy for cooking, air conditioning, home heating, transportation, vehicle refuelling etc., and industry uses energy for production (Godish, 2004). Consequently, these activities of high energy consumption emit a large amount of air pollutants into the atmosphere, bringing serious air quality issues.

Emissions from road transport are especially important and deserve distinctive attention in urban areas. Road transport represents a major source of deterioration of the urban air quality throughout the world (Hoek et al., 2002; EEA, 2012a). Twofold differences in the concentrations of several traffic-related air pollutants in locations with high and low traffic activity have been reported (Martuzevicius et al., 2008). Several findings also suggest that the demand of transportation will exceed improvements related with emission reduction technologies (Delucchi, 2000). Since 1990, some traffic–related air pollutants emissions, such as nitrogen oxides (NOx), carbon monoxide (CO), or non-methane volatile organic compounds (NMVOC) have decreased (EEA, 2010) in European Union, mainly due to the introduction of new technologies (i.e. three way catalytic converters on passenger cars) and stricter regulation of emissions from heavy duty vehicles (Regulation 595/2009). Diesel particulate filter technology was also introduced to mitigate PM emissions. Emission trends compiled for the period 2000–2008 indicate that particulate matter with an equivalent aerodynamic diameter of less than 10 μm (PM10) emissions decreased by 8%, while particulate matter with an equivalent aerodynamic diameter of less than 2.5 μm (PM2.5) was reduced by 13% (EEA, 2010). But in spite of these reductions in air pollutant emissions, the demand for road transport has been growing much faster than anticipated. In Europe, between 1995 and 2010, passenger transport demand by car increased by nearly 21.5%. The car dominates passenger transport mode share accounting for 84% in terms of passenger-km (excluding powered two wheels), followed by bus (9%) and rail

(7%). Also, road transport dominates freight transport mode share with 77% (EEA, 2012a). Road transport remains the most important source of NMVOCs, PM2.5 and PM10 emissions (Figure 1.1) (EEA, 2010; EEA, 2012a). The trends in emissions of PM2.5 have been tempered by the increased market penetration of diesel vehicles since 1990, as also reflected in the final energy consumption by fuel indicator and by the growth in car registrations by fuel type in the EEA (EEA, 2012b).

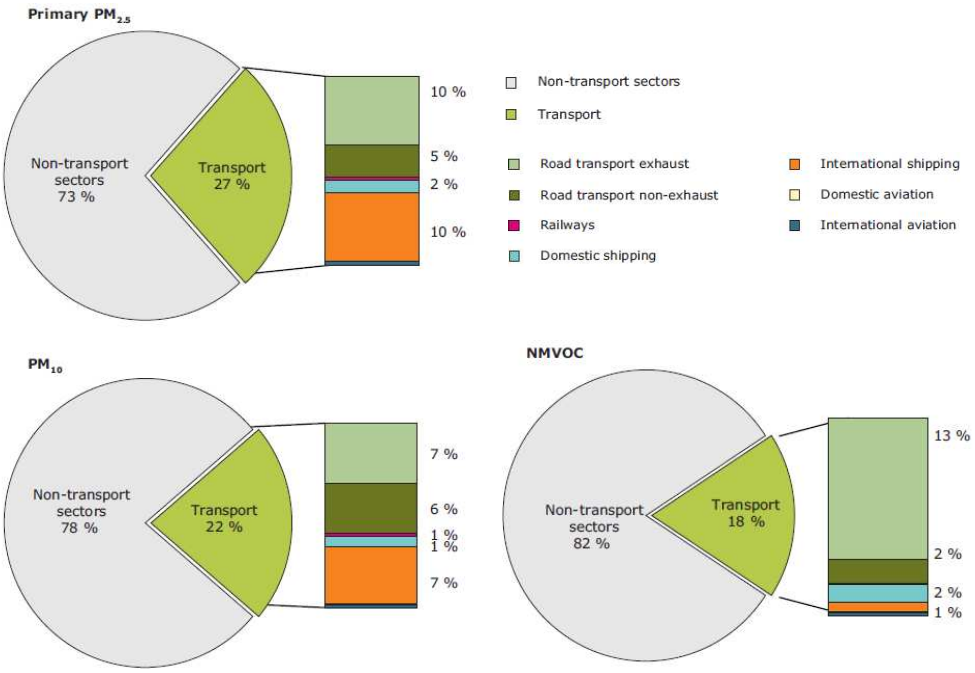

Figure 1.1. The contribution of the road transport sector to emissions of PM10 and PM2.5 in 2010 EEA−32 (EEA, 2010).

Among the extended number of air pollutants emitted by the road transport in urban areas, hazardous air pollutants (HAPs), referred also as air toxics, have been targeted for special attention due to their link with mortality and morbidity at levels usually experienced by individuals in urban areas and the need for action to minimize these risks (Monn, 2001; USEPA, 2007; HEI, 2007; Anderson, 2009; HEI, 2010). Given the toxic and carcinogenic proprieties of such pollutants, a list of 188 HAPs associated with anthropogenic sources was defined in Clean Air Act by the US Environmental Protection Agency (USEPA, 2004a), identifying the benzene, 1,3-butadiene, formaldehyde, acetaldehyde, acrolein, naphthalene and diesel particulate matter (PM) as the major HAPs emitted by mobile sources (USEPA, 2007). Emissions of HAPs are mainly related with incomplete combustion (e.g. benzene) and by-products formed during incomplete combustion (e.g. formaldehyde, acetaldehyde, and 1,3-butadiene), but evaporative processes of fuel components are also important. For benzene, defined as one of the most important health-based European Union priority HAPs (Bruinen de Bruin et al., 2008), the highest outdoor exposures are also likely to occur in during the refuelling at fuel stations and near gasoline fuel stations within urban areas (Wallace, 1996; HEI, 2010).

Currently, the evidence that the human exposure to current and future traffic-related air pollutants within urban areas exerts significant health effects is well established (Pope and Dockery, 2006; Samet and Krewski, 2007; Anderson, 2009; Russell and Brunekreef, 2009; USEPA, 2009a; Brook et al., 2010) and have been widely recognized by both national governments and multilateral development organizations as a threat to urban populations. Thus, climate change may exacerbate existing environmental and health problems. Changes in the temperature, humidity, wind, and precipitation that may follow future climate can deeply impact air quality because of induced changes in the transport, dispersion, and transformation of air pollutants at multiple scales (Bernard et al., 2001; NRC, 2001; Carvalho et al., 2010). The potential impact of climate change on traffic-related air pollution, namely PM, is of a major concern since future changes in their concentrations are likely the most important component of changes in mortalities attributable to air pollution in future scenarios (West et al., 2007).

The European Union has introduced and implemented air quality directives to regulate ambient air quality by setting air pollutant standards and limit values in order to avoid, prevent or reduce harmful effects on human health and the environment as a whole (Directive 2008/50/EC). These directives imply that member states undertake measurements at outdoor locations by fixed-site air quality monitoring networks in order to assess compliance with agreed standard target and limit values that are set with respect to whether short-term or long-term exposure.

Even though the regulatory efforts, such air quality measurements indicates that the Member States of the European Union still have difficulty complying with the legislated limits of traffic related pollutants (EEA, 2012a). In the period 2001–2010, 18 – 41% of the Europe's urban population was potentially exposed to ambient concentrations of PM10 above the EU daily limit value set for the protection of human health (i.e. a daily average concentration of 50 $\mu g.m^{-3}$ cannot be exceeded more than 35 days per year). Moreover, in 2010 the PM10 daily limit value was exceeded at 33% of the traffic stations and 29% of urban background stations within the EU (Figure 1.2). These figures have increased for traffic locations compared to 2009 (EEA, 2012b). For PM2.5, its annual target value (25 $\mu g.m^{-3}$) was exceeded at 6% of traffic sites and 14% of urban background sites. In the case of benzene, except at four stations, measured concentrations in Europe are well below the limit value (annual average concentration of 5 $\mu g.m^{-3}$). However, it should be mentioned that benzene starts to be measured recently at a relatively small number of stations in Europe. Therefore, information on their spatial and temporal variation is limited.

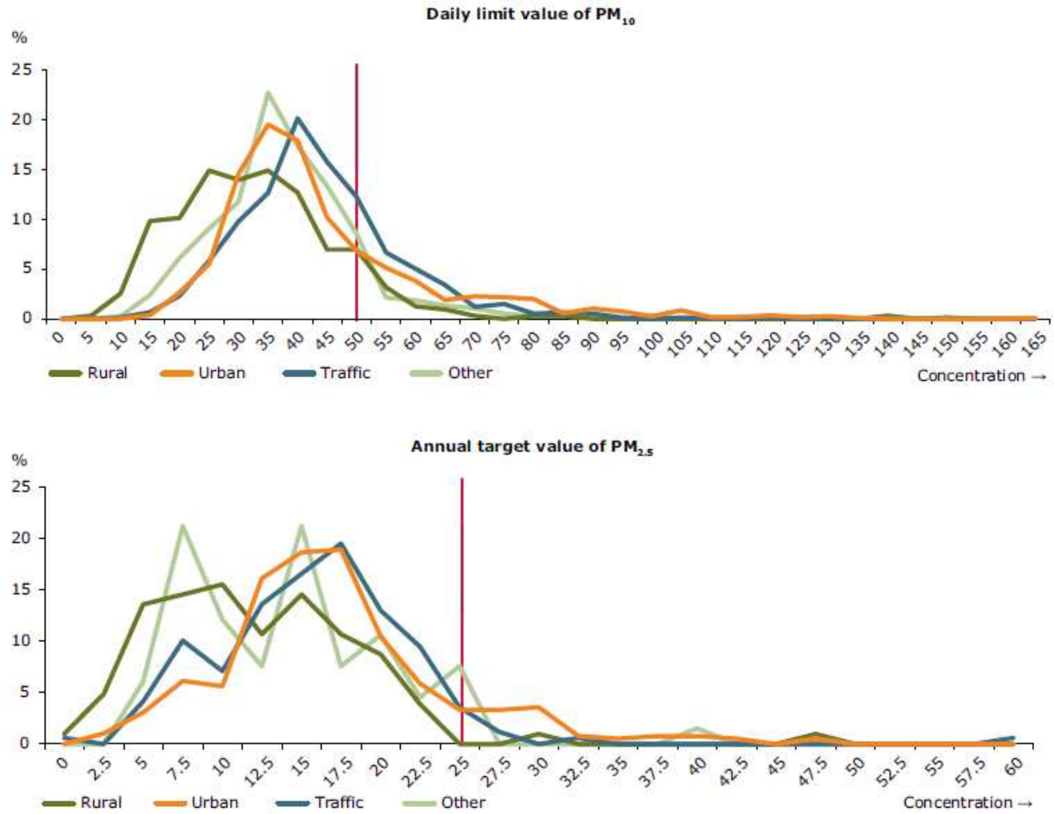

Figure 1.2. Percentage frequency distribution of stations in the EU Member States versus the various concentration classes of PM10 and PM2.5 in 2010 (EEA, 2012c).

Overall, road transport has become the dominant source to outdoor air pollution in urban areas and besides the current EU air quality policy framework, many citizens still live in urban areas where air quality limits set for the protection of human health are exceeded, causing premature death and widespread aggravation to health. In order to protect public health it is necessary to reduce the levels of these exposures and to do so adequately a deeper understanding of source-receptor relationship and interaction between exposure and health effects is needed. Characterizing the magnitude of those exposures and quantifying the average exposure burden imposed by living near traffic are among the problems that need to be addressed.

1.1.2. How human exposure to urban air pollution is defined?

A review of the literature in the diverse fields of exposure assessment, environmental policy and management, risk assessment, environmental health, toxicology, and epidemiology reveals inconsistent definitions of "exposure", depending on the needs and objectives of the different research areas. Thus, several researchers discuss exposure as a quantitative measure of the environmental pollutant like "the concentrations of pollutant in the ambient air, soil, food and water" (IPCS, 1994; Landers and Yu, 1995; Moriarty, 1999), or is mentioned as a qualitative measure of the severity of the environmental pollution. Moreover, in epidemiology, according to the book "Principles of Exposure Measurement in Epidemiology", exposure is defined as "any of a subject's attributes or any agent with which he or she may come in contact that may be relevant to his or her health", suggesting that a behaviour, such as smoking, is an exposure (Armstrong et al., 1992; White et al., 2008). Other references define exposure as a "potential cause of disease" (Monson, 1980; Kriebel et al., 2007), or "the opportunity of a susceptible host to acquire an infection by either a direct or indirect mode of transmission" (Lisella, 1994). Besides the diversity of exposure definitions in the scientific literature, some references use the term "exposure" without defining it at all (IPCS, 1994). On the other hand, for human studies, the concentration at the boundary of contact is the most relevant quantity. However, the boundary of contact is not clearly defined, thus contributing to the misunderstanding as to exposure exact meaning (Moschandreas and Saksena, 2002). Also, the word exposure has different meanings in different contexts. The Monitoring Ambient Air Quality for Health Impact Assessment guidelines (WHO, 1999) distinguishes personal exposure and populational exposure, defining thus personal exposure as true integrated concentrations experienced by individuals and states that populational exposure summarizes the exposure of everyone in the population. Under these guidelines, ambient air quality levels can be used as surrogates of personal exposure.

Despite the discrepancies in the use and definitions of exposure-related terms in the diverse fields of exposure assessment, there is a predominant definition of exposure involving the contact between a physical, chemical or biological agent and the organism target (e.g. human) (Duan et al., 1989; Lioy, 1991; Duan and Ott, 1992; Georgopoulos and Lioy, 1994; Nieuwenhuijsen, 2003; USEPA, 2005; Frumkin, 2005). Under this approach, for human exposure to occur it is necessary a contact between the agent and the external boundary of the human body, such as the airways, the skin and the mouth. As to human exposure to air pollution discussed in this study, the breathing zone is considered the most important point of contact, and inhalation is considered the most important pathway of exposure (WHO, 2000; Moschandreas and Saksena, 2002; Klepeis, 2006).

Several references, however, recognized that it was important to address the time interval over which contact occurs in an exposure event for a quantitative definition of exposure (NRC, 1991; USEPA, 1992; Georgopolous and Lioy, 1994; Zartarian et al., 1997; Zartarian et al., 2004), i.e. exposure duration. Under this context, in 1999, the International Programme on Chemical Safety (IPCS) of the World Health Organization (WHO) initiated a Harmonization Project with an Exposure Assessment Planning Workgroup to confront the issues hindering harmonization in the area of exposure assessment (Callahan et al., 2001; Hammerstrom et al., 2002; WHO, 2004). In 2004, the IPCS glossary was adopted as the official glossary of the International Society of Exposure Analysis (ISEA) (Zartarian et al., 2004) defining thus exposure as the "concentration or amount of a particular agent that reaches a target organism, system, or (sub)population in a specific frequency for a defined duration" (WHO, 2004; Van Leeuwen et al., 2007; IPCS, 2009). Recently, the increasing evidence that each individual is subject to his own individual exposure due to his daily activity patterns (Elliott et al., 2000; Monn, 2001; Sexton et al., 2007; Hinwood et al., 2007) highlights that human exposure to air pollution is not a static phenomenon, also making a clear distinction between population exposure and personal exposure. In attempt to focus at individual level, Branis (2010) defines personal exposure to air pollution as the measurement of a pollutant of concern performed by a monitor (or sampler) worn by a person while the sample is taken from a point near the breathing zone of the person.

To characterize human exposure to air pollution, three aspects are also recognized as important: magnitude – *"What is the pollutant concentration?*"; frequency – *"How often?";* and duration of contact – *"For how long?".* The magnitude of exposure is a critical characteristic in determining adverse effects. Similarly, both the frequency and the duration of exposures can have an important impact. Exposure can be continuous, intermittent, cyclic or random depending upon the source of the air pollutant and individual activities that lead to contact with the pollutant. Also, in order to evaluate the real impacts of urban air pollution in the human health it is important to distinguish between short- and long-term exposures because of the differences in their health effects. Thus, the long-term (i.e. years or lifetime) is related to extended time periods of exposure leading to chronic health effects, whereas in the short-term (i.e. minutes to days), high exposure may show acute effect on human beings unless extremely high concentrations are reached.

Exposure is characterized as a function of concentration and time and can be represented by several time exposure metrics. Depending on the time of exposure, instantaneous, time-integrated and time-average exposure could be distinguished (USEPA, 1992; Ott, 1995; Monn, 2001). The instantaneous exposure is the exposure at an instant in time and it is expressed in the same unit as the concentration (e.g. µg.m^{-3}), while the time-integrated exposure is the integral of instantaneous exposures over the duration of exposure (units: ppmh or µg.m^{-3} h) (Equation 1.1). (Lioy, 1990; Zhang and Lioy, 2002).

$$E_i = \int_{t_1}^{t_2} C_i\,(x, y, z, t)\, dt \qquad (1.1)$$

where E_i is the time-integrated exposure experienced by the individual i, C_i (x,y,z,t) is the concentration occurring at a particular point occupied by the individual i at time t and spatial coordinate (x,y,z), corresponding t_1 and t_2 to the starting and ending times of the exposure event, respectively. This type of exposure can be estimated by measurements (e.g. via personal air monitors) that usually provide incremental data on exposure (NRC, 1991; USEPA, 1992).

Other possible formulations of exposure that depend on the time of exposure include time-averaged exposure and peak exposure (units: ppm or µg.m^{-3}) (Armstrong et al., 1992; Ott, 1995; Zhang and Lioy, 2002). Time-averaged exposure is determined by dividing the time-integrated exposure by the duration of the exposure ($t_1 – t_2$) (Equation 1.2). This can be a useful formulation for many environmental applications (e.g. daily average exposure) and is relevant for long-term exposure and chronic health effects. The peak exposure is usually relevant for short-term exposure and acute toxic effects (Duan et al., 1990; Nieuwenhuijsen, 2003).

$$E_i = \frac{1}{t_2 - t_1} \int_{t_1}^{t_2} C_i(x, y, z, t)\, dt \qquad (1.2)$$

A hypothetical exposure time profile or the exposure time-series. i.e. a plot of concentration as a function of time is presented in Figure 1.3 illustrating several time exposure metrics that may be derived from this profile. The time period to consider in the exposure time profile should be defined under the scope of the exposure analysis (e.g. a biologically relevant time period).

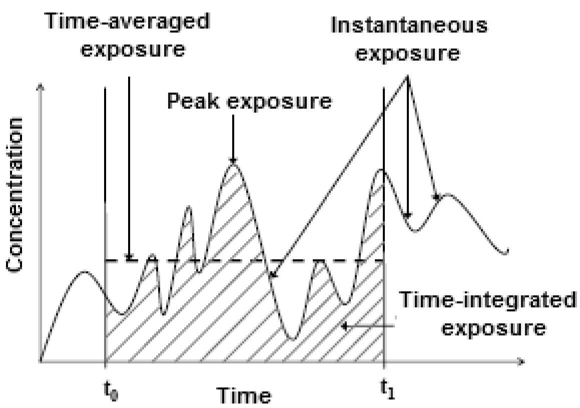

Figure 1.3. Hypothetical exposure time profile and exposure metrics (Duan et al., 1990; Monn, 2001).

In a pragmatic and static approach, the exposure is simply deduced by the air pollutant concentration in ambient (outdoor) air (Sexton and Ryan, 1988; Monn, 2001; Zhang and Lioy, 2002; Özkaynak et al., 2008). However, it is important to mention that there is a clear distinction between the air pollution concentration and exposure concentration. High air pollution concentrations do not necessarily result in high exposures. The concentration of a specific air pollutant is a quantitative expression of the presence of a pollutant in ambient air at a particular place and time (i.e. $\mu g.m^{-3}$ or ppm) and is subject to high variability in space and over time depending on variations of emission sources, meteorology, land use and terrain lead (Zanetti, 2003; Wilson and Zawar-Reza, 2006). Exposure concentration, in turn, requires the simultaneous occurrence of two events: an air pollutant concentration at a particular place and time, and the presence of a person at that place and time (Duan, 1992; Ott, 1995; Zartarian et al., 1997; Zartarian et al., 2004), and is characterized by the spatial and temporal dynamics of air pollution concentrations and time-activity patterns of individuals (Gulliver and Briggs, 2005; Georgopoulos et al., 2009; Son et al., 2010; HEI, 2010; Dons et al., 2011) as discussed in section 1.1.3.

External exposure should also be differentiated from internal exposure. Once the pollutant has crossed a physical boundary (e.g. skin, alveolar epithelial cells) of an individual, the concept of internal exposure is used (WHO, 2000; Ott et al., 2007). Internal exposure is often obtained from biomarkers (Section 1.2.1) as a way of validating cumulative human exposure.

In this context, individual exposure to air pollution is considered in this study as the real concentration of air pollutant breathed in by the individual at a particular time and place, and it does not only arise from the pollutant concentration in the environment through the individual is exposed but is also determined by the amount of time spent in that environment.

1.1.3. What are the needs and the key elements of personal exposure assessment?

Given the well established evidence of causal relationship between human health effects and exposure to air pollution in urban areas (Pope and Dockery, 2006; Samet and Krewski, 2007; Anderson, 2009; Russell and Brunekreef, 2009; USEPA, 2009a; Brook et al., 2010) it is necessary to determine the amounts of air pollutants to which general individuals are actually exposed to assess the impact of air pollution on human health. Thus, human exposure assessment emerged in context of scientific research as an important analysis tool to prevent public health from the harmful effects of air pollution.

Human exposure assessment is an important tool to describe and determine, qualitatively and quantitatively, the pollutants´ contact with the human body (WHO, 2006), and is a critical parameter of epidemiology and health impact assessment (HIA). Epidemiology relies on the inference of associations between exposure and response variables. Typically, the quantitative estimates of exposure-response in epidemiological studies reflect the late-stage end points of morbidity, mortality and tissue pathology

(Kauppinen, 1996; Bocchetta and Carbone, 2004; Maier et al., 2004). Exposure assessment is one of the four major components in the HIA process (Figure 1.4), and also often one of the most demanding. HIA provides the probability, magnitude and uncertainty of health effects associated with exposure.

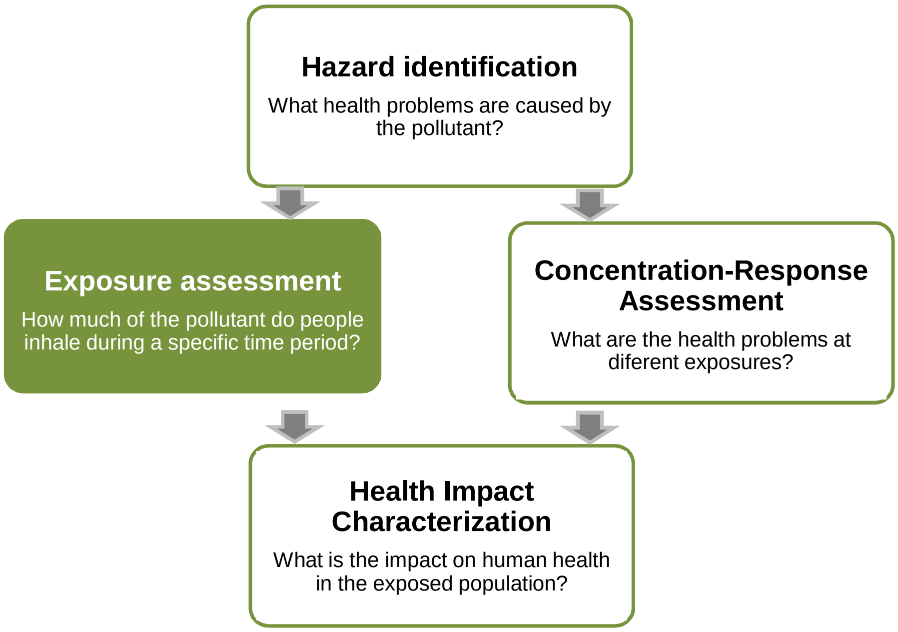

Figure 1.4. Elements of health impact assessment process (USEPA, 2012).

In this prespective, characterizing and estimating the magnitude of potential exposures is an essential component for evaluating the potential health effects posed by a particular pollutant (Moschandreas and Saksena, 2002). The potential effects on human health can be quantified based on the number of cases attributable to air pollution that may be prevented by reducing current levels of air pollution (Künzli et al., 2000), as presented in Figure 1.5. An estimate of attributable deaths (AD) is obtained from the average number of deaths, the exposure-response function and the regression coefficient β provided by epidemiological studies that characterise the ratio for a unit increase in pollutant concentration, and the difference between the daily average concentration (x) and a reference value under given scenario (x_0).

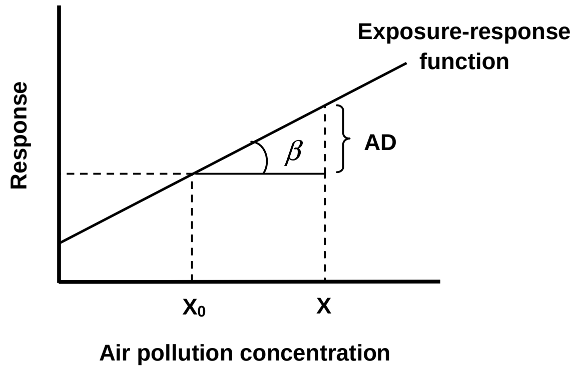

Figure 1.5. Methodology to derive number of cases attributable to air pollution (Künzli et al., 2000).

The science of human exposure assessment has become substantially more complex over the past decades as the demand for relevant and accurate human exposure information has increased in all the scientific fields related to public health protection. Over the past 20 years, numerous methods for assessing human exposure levels to air pollution have been used by several studies focusing on the links between air pollution and health, with the ultimate goal of estimating exposure at individual level within an entire study population (Lebret et al., 2000; Kousa et al., 2002; Arteta et al., 2006). However, the main criticism of these studies relates to the quality of the air pollution exposure, leading to inaccuracies and underestimation of the health impacts (Weis et al., 2005; Szpiro et al., 2008; Nuckols et al., 2010; Peng and Bell, 2010).

In this context, taking into account the source-to-outcome framework developed by the National Research Council (NRC, 1998), the processes that are important for exposure science start with a pollutant entering the environment and end with health effect characterization. This framework includes two steps focused on the place (pollutant source emission and pollutant dispersion and transformation), while the third step focuses primarily on human being (human exposure and adverse health effects). Despite exposure assessment has made the most significant improvement in quality over the past 20-year history of the HIA, admittedly, there are several key elements that should be considered for personal exposure assessment to capture the spatial and temporal variability of personal exposure to air pollutants in urban areas (Briggs, 2008; Nuckols et al., 2010; de Nazelle et al., 2011), as described below.

◢ Spatial and temporal variability of road transport-related air pollution

Transport emissions are non-homogeneously distributed in space and in time and, therefore contribute to the intra-urban variation in the concentrations of air pollutants. Several specific features of the traffic can be identified as influencing the amount of emissions attributable to road transport and affecting consequently urban air quality (Gwilliam, 2003; WHO, 2005a; EEA, 2012b). They include significant number of vehicles circulating in urban areas, the age of the vehicle fleet and the technology used, the physical characteristics and chemical compositions of fuels and driving conditions. Thus, transport activity represents one of the main input data to estimate road transport emissions. This detailed information can be provided by automatic measurements systems or from transport modelling (André et al., 1999; Boulter et al., 2007). Usually, since it is not possible to obtain enough measurements for the entire study area with the resolution required, transportation models represent a consistent approach to characterize transport activity within urban areas, providing detailed information on traffic flow for each road segment. Also, it is possible to distinguish between different vehicles categories, such as private passenger cars, public transport, goods transport etc., while automatic measurement systems usually provide only the total number of vehicles. Another important characteristic for the transport sector directly related with atmospheric pollution is the average age of vehicles. Older vehicles are associated with higher emissions of air pollutants than newer vehicles, because performance deteriorates as a function of age and older vehicles are more unlikely to use emission reduction technology. In addition, the congested urban traffic conditions and large number of short trips can result in higher emissions per kilometre.

Currently, several methodologies to quantify the pollutant amount emitted by the vehicles to the atmosphere are available. They range from calculations at microscopic scale (i.e. for a single vehicle, or for a street) to macroscopic calculation (i.e. regional, national and global levels) (Joumard, 1999; Agostini et al., 2005; Gkatzoflias et al., 2007; Smit et al., 2007). However, the modelling tools not always cover HAPs or provide emissions with low temporal and spatial resolution that is not sufficient for urban scale studies. To be applied for the urban areas, the currently existing methodologies of the emission quantification have to be adapted taking into account availability of the input data and final use of the emission estimation results. Thus, due to importance of such requirements in this research, a new version of the available Transport emission Model for Line Sources (TREM) has been developed for HAPs providing detailed information concerning traffic emissions for each road segment in urban areas (Tchepel et al., 2012).

After the releasing of air pollutants into urban environment by emission sources, they can be transported and transformed through a number of physical and chemical processes at a range of spatial and temporal scales (Figure 1.6). At scales ranging from a simple building and street canyons to the entire city, microscale mechanical and thermally driven turbulence dominates local dispersion processes. However, these processes operate within a hierarchy of larger scales which provide the background state of the atmosphere that modulates air quality within urban areas (Wilson and Zawar-Reza, 2006;

Solomon et al., 2008; Turner and Allen, 2008). In focusing on air quality in the urban atmosphere, the emission source activity and weather or topological conditions will significantly affect the spatial and temporal variation of the ambient concentrations in the urban environment, influencing thus the personal exposure to air pollution depending on when and where people spend their time.

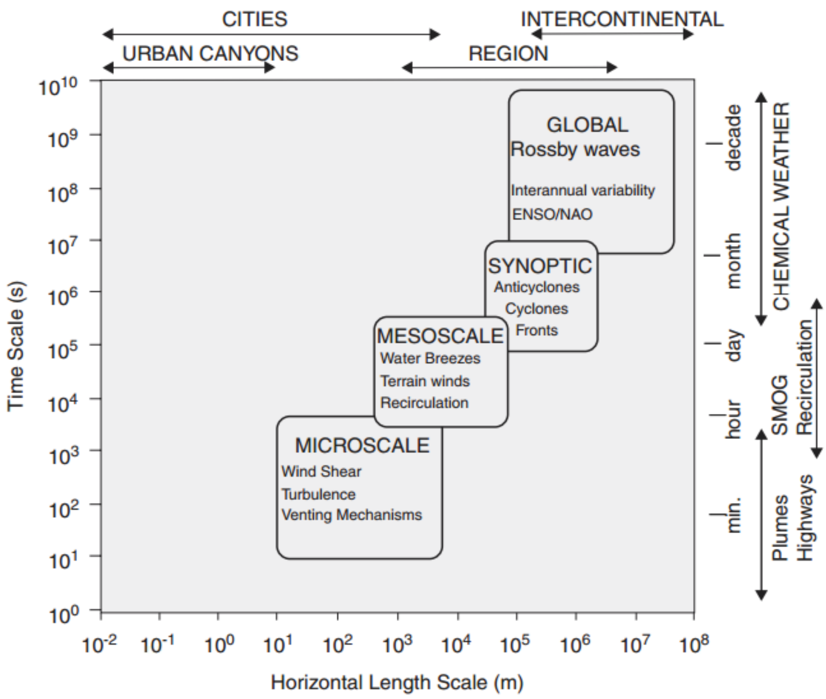

Figure 1.6. Temporal and spatial scales affecting atmospheric dispersion in the urban environment (Salmond and Mckendry, 2009).

In urban areas, the transport and dilution of air pollutants are affected by meteorological conditions and physical structures of the city. The presence of high buildings on either side of the road creates a “street canyon”, which reduces the dispersion of the emitted pollutants from traffic sources and can lead to significantly higher concentrations locally. There is also evidence to suggest that air pollution concentrations fall virtually to background levels behind a row of uninterrupted buildings (Bloemen et al., 1993). Various monitoring studies have suggested that in cities, strong variability of traffic-related air pollution may occur over small distances (<100 m) (Monn et al., 1997; Roorda-Knape et al., 1998; Nikolova et al., 2011), so that the pollution data from a single fixed-monitoring site can only be considered representative of a rather small surrounding area.

In this concern, to analyse the high spatial and temporal variability of road transport-related air pollution within the urban environment where inhabitants are living close to the pollution sources, it is required to characterize the transport activity in order to quantify the corresponding emissions and air pollutants levels. For this purpose, a system based on the transportation modelling linked with the emissions and dispersion modelling is considered as one of the most suitable approaches to provide detailed information concerning traffic flow for each road segment and related pollution (Borrego et al., 2006).

◢ Contribution of indoor concentrations

Urban air is an umbrella concept, combining outdoor and indoor air. Additionally to significant temporal and spatial variability of outdoor concentrations, scientific evidence has shown that indoor environment plays a significant role in personal exposure to air pollution, where urban populations spend large fractions of their time throughout life (Koistinen et al., 2001; Baklanov et al., 2007; Georgopoulos et al., 2009; Zou et al., 2009a). Thus, indoor spaces represent important microenvironments when addressing health effects from air pollution.

Indeed, human exposure should not be associated exclusively with outdoor air. Several studies on exposure noted that using only the outdoor component of exposure is not sufficient as several potentially confounding variables are omitted from the exposure assessment process (Lioy, 1990; Monn et al., 1997; Boudet et al., 2000). In this sense, the contribution of indoor air to personal exposure has been increasingly recognized as being of importance (Wallace, 1996; Jantunen and Jaakkola, 1997; Samet and Spengler, 2003; Adgate et al., 2004a, 2004b; Phillips et al., 2005; Mitchel et al., 2007; Colbeck and Nasir, 2010). Also, it is known that most people in European urban areas spend 80–90% of their time indoors during the average day, 1–7% in vehicle, and only 2–7% outdoors (Colls, 2002; Brunekreef et al., 2005; Koutrakis et al., 2005). Despite the research community recognize its importance, policy makers have focused their attention on outdoor air quality and non-occupational air pollution regulations have typically been applied focusing on outdoor rather than indoor air. For this purpose, observations from stationary outdoor monitoring sites are usually considered, which means that air pollutants from indoor sources have been ignored.

Nevertheless, several findings indicate that indoor concentrations are typically higher than the respective ambient levels (Figure 1.7). Also, in case of benzene have been consistently demonstrated that its concentrations tend to be higher in the colder than the warmer seasons (Edwards and Jantunen, 2001; Schneider et al., 2001; Amagai et al., 2002). For formaldehyde, indoor exposures are also the dominant contributor to personal exposures through inhalation, corresponding to about 98%, and indoor concentrations may be high enough to cause adverse health effects (EC, 2005). In addition, Wilson and Suh

(1997) conducted a meta-analysis of data from multiple sites and concluded that concentrations of fine particles originating from indoor sources are weakly related with ambient levels over time.

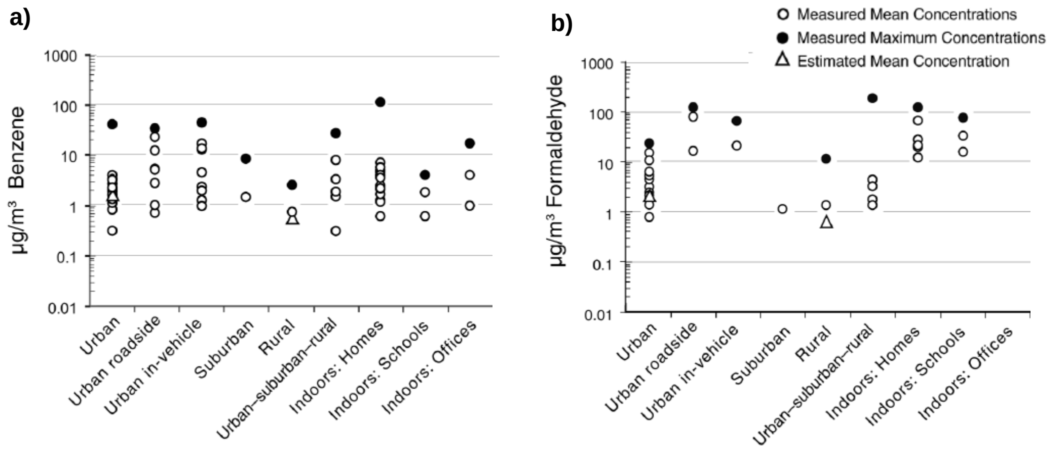

Figure 1.7. Range of mean and maximum concentrations (μg.m^{-3}) of a) benzene and b) formaldehyde, at various indoor and outdoor locations (HEI, 2007).

Under this framework, individual exposure to air pollution depends greatly on indoor concentrations, which in turn vary widely between indoor spaces as a function of location and time. The extent of these variations depends on a set of factors including their indoor emissions, mixing with infiltrated outdoor air, ventilation conditions and occupant behaviour. Also, the diffusion of outdoor air into buildings contributes to a mixture of indoor and outdoor pollutants and resulting indoor exposure levels according to several factors (e.g. air conditioning and the indoor–outdoor temperature gradient) (Lai et al., 2004; Branis, 2010). Traffic-related air pollutants generated from outdoor sources, such as PM2.5 both effectively penetrate and persist in many indoor environments. Indoor environments also present a variety of emission sources which are independent of the outdoor environment, such as cooking, environmental tobacco smoke, burning of natural gas or wood, building materials (e.g. polyurethane foams), furnishings and certain consumer products (e.g. adhesives).

Actually, when comparing with road transport emissions, the contributions of indoor sources are generally small but with sharp presence during the time-activity patterns of individuals, modifying individual's exposure substantially (Rodes et al., 1991; Freeman and Saenz de Tejada, 2002; Ferro et al., 2004; WHO, 2005a; Franklin, 2007). Roorda-Knape et al. (1998) reported an average concentration of 91.6 μg.m^{-3} for PM10 in 11 schools located near highways. The authors pointed out, however, that indoor concentrations of PM10 were largely controlled by indoor activities of the occupants rather than by traffic. Thus, measurements performed at 4 schools in Viseu within the framework of Portuguese national project SaudAr demonstrated that indoor levels of PM at schools were higher than outdoors during the working days and low indoor levels observed during weekends suggested that higher PM concentration are related to human activities (Valente, 2010). In case of formaldehyde, Pegas et al. (2011) reported that indoor concentrations in three schools in Lisbon were markedly higher than those observed outdoors. Higher levels in classrooms than outdoors suggest that indoor sources are more important contributors to the indoor levels than outdoor sources, such as infiltration of vehicle exhaust. Nevertheless, it is important to note that sampling indoor air is not enough to understand personal exposure and has been demonstrated that personal exposure does not correlate well with measurements of indoor concentrations (Monn, 2001).

◢ Importance of time-activity patterns

The understanding of human behaviour during daily life is a topic of interest within several social sciences. Human behaviour and use of time is referred to as the time-activity pattern of an individual, and are strongly linked to various personal characteristics including age, gender, education, income and employment status (Pas, 1984). During the course of their daily activities, air pollution levels, changing dramatically in space and time, influence an individual's exposures. Thus, time-activity patterns play a significant role, if not the most significant role, in characterizing personal exposure (McKone et al., 2008).

Urban areas, where currently lives around 75% of the European population, are a complex systems with individuals characterized by different behavioural patterns (Galea and Vlahov, 2005; Batty, 2009; Portugali et al., 2012). For decades, urban spatial structure measured by the degree of spatial distribution of population and employment, has been studied to describe the structure and organization of cities, and their function and role in people's life (Horton and Reynolds, 1971; Anas et al., 1998; Florida et al., 2008). Cities supply individuals with resources as well as constraints. The urban environment accommodates services, employment opportunities and other facilities where individuals may conduct desired activities, affecting significantly their mobility.

One of the earliest spatially integrated perspectives for the analysis of time-activity patterns and movement in space and time is time geography. Time geography rests on the notion that the locations and movements of individuals can be followed and visualized as continuous paths in spatial and temporal dimensions (Figure 1.8). A time geographical approach allows for the examination of place as the spatial, temporal and contextual terrains that influence individual health status (Thrift, 1977; Parkes and Thrift, 1980; Miller, 2001; Miller, 2007).

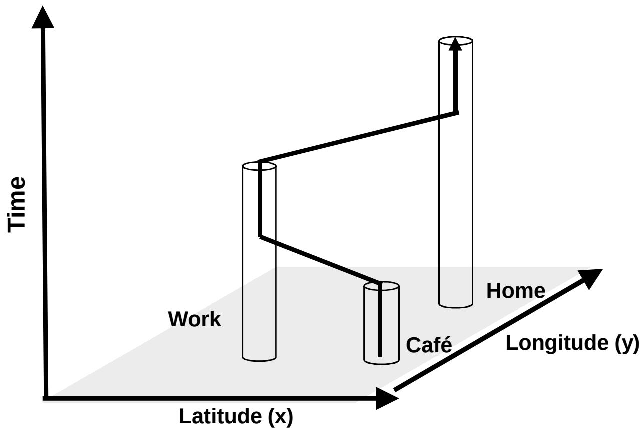

Figure 1.8. Time-activity patterns of an individual (Miller, 2007).

In the context of human exposure, an understanding of human mobility patterns is crucial as they strongly influence the assessment accuracy of actual human exposure to air pollution (Harrison et al., 2002; Nuckols et al., 2004; WHO, 2005b; Nethery et al., 2008; Beckx et al., 2009; Dons et al., 2011). Analysing time-activity patterns for personal exposure assessment may indicate the distribution of time among activities and the factors that influence the degree of media contamination in the activities, and reflect the duration of contact during the activities (Zou et al., 2009b). Also, there is an inter and intra-variability of individual's activities, which has implications for the use of time-activity data in exposure assessment. A review of studies on time–activity patterns used in epidemiologic studies is given by Ackermann-Liebrich et al. (1995). The information needed in such studies include location of the activity, the period of time when the activity took place (e.g. time of day, phase in life), and the duration of the activity.

International and national studies focusing on human exposure to air pollution, such as TEAM studies (Wallace, 1991), the National Human Exposure Assessment Survey (NHEXAS) (Freeman et al., 1999), the National Human Activity Pattern Survey (NHAPS) (Klepeis et al., 2001), the Population Exposure to Air Pollutants in Europe (PEOPLE) project (Ballesta et al., 2006), the Health and The Air We Breathe (SaudAr) project (Borrego et al., 2008; Valente et al., 2008) or the Air Quality Exposure and Human Health in Industrialized Urban Areas (INSPIRAR) project (PTDC/AAC-AMB/103895/2008, ongoing project) were relying on diary-based instruments (e.g. time-activity diary (TAD), questionnaires, etc.) to categorize the environments where exposure occurred and sources of air pollutants, and to derive information on the temporal sequencing of human activities during the study period. However, such time-activity information does not account for the movement of individual and mostly lacks the exact “activity-space” where a specific activity is executed by the individual (Harvey and Pentland, 1999; Rainham et al., 2010; Lawless et al., 2012), and the sequence of exposure events is not considered. Thus, problems in quantifying personal exposure still remain. By using approximations for exposure, health effects can be wrongly assigned, or the strength of a relationship will not be sufficiently emphasized (Jerrett et al., 2005b; Piechocki-Minguy et al., 2006; Dons et al., 2011; Physick et al., 2011; Setton et al., 2011).

Also, home addresses are generally used as the surrogate for the personal exposure, when in fact a high percentage of an individual's exposure can accrue from relatively short periods of time spent in high-polluted indoor environments (Harrison et al., 2002; Nethery et al., 2008; HEI, 2010; Dons et al., 2011). This suggests that spatial variations and fluctuations over time imply that even two individuals living in the same residence are subject to their own individual exposure due to their time-activity patterns (Elliott et al., 2000; Monn, 2001; Sexton et al., 2007; Hinwood et al., 2007). In addition, several findings indicate that the time spent at workplace greatly contribute to within-area exposure variability (Setton et al., 2008) and may also substantially increase exposure, compared with the data at fixed monitoring sites (Baklanov et al., 2007). Thus, to overcome some of the difficulties inherent to the collection of time-activity information, new technologies, such as global positioning system (GPS), and related activity-measuring devices, such as accelerometers, offer possibilities for reducing such errors in the exposure assignment of individuals in health studies (Section 1.2.2).

Summary 1.1.: *Currently, many citizens in urban areas are exposed to air pollution levels that exceed the air quality limits set by the legislation for the protection of human health, with road transport being the most significant pollution source. Among the extended pollutants emitted by road transport, hazardous air pollutants require special attention due to their link with cancer and other serious adverse effects on human health. Thus, personal exposure estimation is crucial to determine the relationship between the air pollution and health effects, and is the most accurate indicator of what individual actually breathe, arising not only from the pollutant concentration in the environment but also depends on the amount of time spent by the individual in that environment. The poor correlations often observed between individual exposures and fixed-site ambient air concentrations suggest that a set of factors other than ambient air may contribute to personal exposures. The spatial and temporal variability of air pollutants in combination with indoor exposures and time-activity patterns are key elements to a proper assessment of personal exposure to air pollution in urban areas and subsequent health effects. The large variability imposed by all these key factors causes* *individual exposure to be a highly dynamic process rather than a static phenomenon, and consequently, an individual's personal air pollution scenario can be very unique*, *thus emphasising the importance and need for personal exposure assessment. Thus, it is clear that analysing individual exposure in urban areas offers several challenges where both individuals and air pollution levels demonstrate a large degree of variability over space and time. Despite time–activity studies have illuminated relatively consistent patterns of activity between different populations, these studies have not enough investigated the crucial question of ''where'' individuals are the rest of the time.*

1.2. Personal exposure assessment: methods and advanced technologies

During the last decades, several exposure assessment approaches have emerged on the exposure research field. However, given the emergence of new technologies to understand the relationship between the environment and individuals, the need to move beyond a static perspective in exposure assessment to include a dynamic approach is evident. Thus, quantifying the contribution of human exposure with observed health symptoms presents further challenges in urban areas.

1.2.1. How personal exposure to air pollution can be quantified?

On a traditional approach, the evaluation of human exposure to air pollution can be carried out under a (i) direct approach or (ii) indirect approach. On a direct approach, exposure levels are measured at the individual, based on personal monitoring or using biological markers (Grandjean, 1995; Lioy, 1995) while under an indirect approach, exposure levels are usually estimated or modelled based on ambient measurements, exposure modelling and surveys (Monn, 2001). In addition, a review of the literature also reveals other frameworks classification such as (i) point-of-contact measurement or personal monitoring in which exposure can be measured at the point of contact (the external boundary of the body) while it is taking place, (ii) reconstruction of internal exposure, which exposure in turn can be reconstructed through internal indicators (biomarkers, body burden, excretion levels, etc.) after the exposure has taken place and also (iii) exposure scenario evaluation in which the exposure is estimated considering hypothetical but plausible scenarios to analyse the concentration and contact time, including the application of models (USEPA, 1992; Callahan and Bryan, 1994; Lioy, 1995).

Nevertheless, under a traditional framework, major air pollution exposure assessment are based on a static perspective assuming a static place/ location for the individual and emphasize population exposure assessment rather than individual exposure assessment. However, individual exposure to air pollution in urban areas results from dynamic process and multifaceted iterations between the human being and urban air. Analysing and refining the understanding of the relationships between people, place, and human activities have been identified as important priorities in several research fields, particularly for research on health and environment (Miller, 2007; Matthews, 2011). For example, on the perspective of human-environment geography two different groups of research methods exist: *place-based approaches* and *people-based approaches* (Miller, 2007). Place-based approaches reflect human-environment interaction at certain locations without considering human beings' activities thoroughly. By contrast, people-based approaches focuses on human beings' activities at a given time and place considering individual's daily activities and their interaction with environment in detail (Miller, 2007).

Currently, new conceptualization of exposure assessment and context that takes the spatial and temporal configuration of exposure has emerged strongly supported by the recent development of geo-spatial technologies (Kwan, 2009; Fang and Lu, 2012; Steinle et al., 2013) and moving thus from a static assessment to dynamic personal exposure assessment. Consequently, beyond the direct approach or indirect approach, personal exposure assessment can also be characterized by two new main groups of methods: *dynamic personal exposure approach* and *static personal exposure approach.* Dynamic personal exposure approach includes the direct methods described above and also spatio-temporally explicit exposure modelling. The static personal exposure approach is related with indirect methods. Both approaches present different exposure estimates, diverging also in relation to precision, costs, viability, and others factor, as following discussed.

◢ Static personal exposure approach

Under a static personal exposure approach, the human activities are considered as a static phenomenon. This approach examines personal exposure to air pollution by subdividing a study area into homogeneous objects (Benenson and Torrens, 2004), based usually on census units or other predefined city boundaries and ambient air quality values obtained by measurement or modelling are considered as surrogate to exposure concentration for each sub-region at a specific time (Zou et al., 2009a). This approach includes fixed-site measurements, surveys, and modelling methods where time-activity patterns of individuals are not directly addressed.

Exposure to air pollution has been traditionally assessed based on ambient air quality measurements provided by fixed-site air quality monitoring networks and based on aggregated demographic data (Rodes et al., 1991; Charpin et al., 1999; Nerriere et al., 2005; Kaur et al., 2007; Sarnat et al., 2009). Thus, in these studies, the same pollution concentration is assigned to people living in defined areas (e.g. city, urban agglomeration). Ambient monitoring networks have been established all over Europe by national institutions. They are equipped with monitors providing continuous data with sufficient time resolution. Monitoring ambient air quality is essential to understanding how the quality of air is changing over time and, in some cases over space, and is an essential tool in managing the environmental impact from air pollution to health and ecosystems (as presented in Section 1.1.1).

Nevertheless, since ambient air monitoring data from a single or few points are unlikely to adequately capture the greater spatial heterogeneity of air pollutants directly emitted from traffic (Kinney et al., 2000; Zhu et al., 2002; Wilson et al., 2005; Zhou and Levy, 2007; Baxter et al., 2013), the issue of considering fixed-monitoring air quality data to human exposure has been analysed (Brauer et al., 2003; Gulliver and Briggs, 2011; Merbitz et al., 2012). Several studies have already examined the correlation between personal exposure and concentrations measured at fixed monitoring stations (Boudet et al., 2001; Gulliver and Briggs, 2005; Baxter et al., 2013). Epidemiological studies within a city consistently find positive associations between outdoor concentrations and health effects due to the high correlation between mean population exposures and outdoor concentrations over time (Janssen et al., 1999; Yip et al., 2004). However, correlations between individuals' personal exposures and their residential outdoor concentration are often weaker, and this may explain the weak associations found in some epidemiological studies (Koutrakis et al., 2005; Sarnat et al., 2006; Van Roosbroeck et al., 2008). Also, results showed that the sampling at the fixed monitoring site may under- or over-estimate air pollutant levels in a ''hot spot'' area, suggesting detailed characterization of spatial distribution of air pollutants for conducting accurate assessment for peak personal exposure (Wu et al., 2005; Ferreira, 2007; Zhu et al., 2008; HEI, 2010). Further, health effects seem to be underestimated when using citywide concentration levels in situations with a high variability in pollution concentrations (Jerrett et al., 2005a; Miller et al., 2007). In this context, fixed-site measurements should be used carefully for personal exposure quantification since they cannot provide good estimates of individual exposure (Brauer et al., 2003; Singh and Sioutas, 2004; Özkaynak et al., 2008; Dons et al., 2011; Merbitz et al., 2012; Baxter et al., 2013).

Another example of static approach for estimating exposure is based spatial surrogates. Spatial surrogates are considered to allocate geographically distributed data to higher resolution geographic areas based on some form of activity or socio-economic/ demographic data (Boulton et al., 2002). This approach is often combined to modelling methods as discussed in Section 1.3. For instance, to assess the exposure to traffic-related air pollution the proximity to traffic or some additional indicator such as composition or volume of traffic is used (Venn et al., 2005; Ryan et al., 2007a). In addition, questionnaires can be used to assess the perception of traffic near the home, representing a surrogate for the traffic intensity and therefore pollution levels in air (Monn, 2001). Questionnaires can also be used to provide information on the existence of exposure sources and to categorize exposure, for example in personal exposure to environmental tobacco smoke (Franklin et al., 1999). The advantage of surrogate data are that they require no actual data on pollution, emissions or meteorology and can therefore be very cheap to collect. However, also these factors constitute the main disadvantage of this method that can be inaccurate, unless well validated.

Overall, the main problem of studies that assess personal exposure on static perspective is that is now widely acknowledge the significant variation of air pollution within urban areas, with the intra-urban variation often greater than inter-urban variation (Jerrett et al., 2005a; Wilson et al., 2006). Thus, the hypothesis on homogeneous air pollution concentration region considered by place-based methods is problematic. Also, the spatial and temporal resolutions are coarse, considering daily, monthly, and even quarterly intervals as time spans (Samet et al., 2000). Air quality modelling, in turn, is a useful tool to overcome this issue, since it provides air quality information and its spatial and temporal variability on a given study area as discussed in Section 1.3.1. Finally, it can be very inaccurate to assume that different individuals in the same region have the identical air pollution exposure level.

◢ **Dynamic personal exposure approach**

A dynamic personal exposure approach assesses human exposure to air pollution at the individual level and takes into account individual activities in space and time. Thus, it considers both individual time-activity patterns and air pollution concentration variability. This approach could estimate personal exposure based on direct methods, such as personal monitoring and biological monitoring, and also spatio-temporally explicit exposure modelling (discussed in detail in Section 1.3.2).

Several exposure analysts believe that personal monitoring is the most reliable and accurate way of estimating the air an individual is actually exposed to (Flachsbart, 2007). Personal monitoring approach assesses an individuals' exposure based on measuring the concentration of a pollutant ideally within a person's breathing zone for a defined time. A variety of active (i.e. pumped instruments) and passive devices (e.g. diffusion tubes) have been used to monitor personal exposure to air pollution as closely as possible to the breathing zone providing the most accurate information about the actual exposure variability (Elliott et al., 2000). Personal exposure monitors collect real-time and time integrated measurements of acute and chronic exposure, respectively. These devices can be either integrating or fast response instruments. Integrating (also called pre-concentration) monitoring techniques collect gaseous pollutants or particles on an appropriate adsorbent bed or filter, respectively, which can be analysed or weighted later in a laboratory. Fast response monitoring may rely on optical or electrochemical techniques to record pollutant concentrations at very high temporal resolution (e.g. one second). Integrating monitoring has been commonly used in personal exposure studies, while fast response instruments are now becoming more popular (Monn, 2001; Branis, 2010; Dons et al., 2013).

Personal monitors should be portable, flexible, robust and user friendly, as well as lightweight and battery operated (or passive) (Nieuwenhuijsen, 2000; Monn, 2001; Branis, 2010). Suitable personal monitors must also fulfil several requirements, such as detection limits, interferences, time resolution, easy operation and cost (ACGIH, 1995; WHO, 2000). Passive air samplers are probably the most convenient tool for conducting large-scale personal exposure assessments (Zabiegała et al., 2010; Król et al., 2012). This is due to the fact that passive samplers do not require a power supply, which in turn means that electrical devices (e.g. pumps), are small, inexpensive and easy to use. However, there is strong dependence of passive sampler performance on meteorological conditions (WHO, 2000; Król et al., 2012), the ability to only record time-integrated concentrations and absorbing capacity is limited (Branis, 2010). For an accurate personal exposure assessment by active samplers, the sampling rate, breakthrough volume and detection limit are important parameters which need to be considered (WHO, 2000).

The personal monitoring is gaining popularity, mainly given the recent technological advances that have reduced the size/weight of personal air samplers while improving accuracy and efficiency. The strength of personal sampling is its provision of real exposure values for the individuals followed. The drawback of this approach, however, is the high cost of implementation. Also, the temporal resolution is limited since this approach provides only exposure data for the individual at the time of sampling, thus limiting the usefulness of its value in estimating long-term exposure. In addition, poor compliance with personal sampler wearing protocols can create positive or negative biases in the reported exposure concentrations, depending on proximity of the participant or the personal sampler to the pollutant source when the monitor was not worn as instructed. This may lead to significant exposure uncertainty related to health inputs in risk assessments (Lawless et al., 2012).

Personal monitoring data serves also as input to and for the validation of exposure models (Hertel et al., 2001; Gulliver and Briggs, 2005; Gerharz et al., 2009; HEI, 2010; Dons et al., 2011).

Also focusing at the individual level, biological monitoring is an emerging tool in the field of personal exposure assessment. It is important to highlight that biological monitoring has been used by epidemiological studies applied to a select group of individuals, i.e. cohorts, who have one or several common characteristics (e.g. gender, age, non-smokers, etc.) to assess internal exposure and health outcomes of individuals during follow-up study, or during their lifetime. Biomonitoring is a direct method for estimating human exposure to air pollutants which accumulate in certain parts of the body, or generate a range of biochemical and physiological responses. Biological monitoring has been increasingly viewed as a desirable alternative to characterize personal exposures not only because it accounts for all possible exposure routes but also because it covers unexpected or accidental exposures and reflects inter-individual differences in uptake or genetic susceptibility (Lin et al., 2002). Biological monitoring refers to measurements of concentrations of biological markers (biomarkers) in human fluids and/or tissues (such as blood, urine, breast milk or hair) to detect exposure. Biological monitoring is a valid tool to provide a direct estimate of internal exposure to a chemical in the individual, which in turn reflects an interaction between an environmental agent and a biological system (Clewell et al., 2008). Collection of biomarkers can be either invasive (e.g. blood sampling) or non-invasive (e.g. urine sampling).

Several studies utilizing biomarkers to assess personal exposure to traffic-related air pollution have been conducted until now (Buckley et al., 1995; DeCaprio, 1997; Scherer et al., 1999; Scherer et al., 2000; Sørensen et al., 2003; Fanou et al., 2006; Hu et al., 2006; Adetona et al., 2013; Baxter et al., 2013). However, the use of biomarkers is most extensive in occupational studies because the exposure–response relationship between pollutants concentrations (e.g. benzene) in such exposures and biomarkers are of importance (Jacob et al., 2007). Biomarkers have been presenting a potential value as proxy measures of disease outcome, and as means of distinguishing individuals who may be unusually susceptible to the effects of a pollutant (Ryan et al., 2007b). Also, several studies have used biomarker analysis to calibrate and to validate the reliability of other exposure estimates (Hertel et al., 2001; Paustenbach and Galbraith, 2006).

Biological monitoring can be used as direct measurements of important individual internal exposure events and to estimate biological effect if a relationship has been established between the biological measurement and the individual health outcome. Thus, biomonitoring presents several strengths for personal exposure assessment to air pollution and can improve the accuracy of exposure assessment. The main advantage of biomonitoring is that only the contaminants that enter the human body are measured. Furthermore, it helps to estimate aggregate exposure, as all exposure pathways are included (inhalation, dermal contact and ingestion), which reflects the comprehensive effect of multiple chemical mixtures, absorbed by all exposure routes, not just air (Monn, 2001). This is, on the other hand, also a limitation as it is not easy to differentiate the component ratios between exposure sources, pathways (e.g. dietary) and chemicals (Ryan et al., 2007b; Clewell et al., 2008). Another constraint is that biomonitoring data may depend on the moment in time when the sample is collected. Depending on the kinetics of the measured compound in the sampled tissue, the measurement may reflect recent exposure, average exposure over a prolonged period of time, or neither (Clewell et al., 2008).

Exposure modelling had arising as an alternative method of dynamic personal exposure assessment able to address the magnitude of air pollutant concentration really breathed in by the individual, allowing to analyse the contribution of different air pollutants, exposure sources and pathways in exposure assessment process (Jerrett et al., 2005a; McKone et al., 2008; Setton et al., 2011; Steinle et al., 2013). Exposure models can be used to investigate large populations, future exposures, as well as reconstruct historical exposure by utilizing existing data from different types and sources, as discussed in detail in Section 1.3. Moreover, exposure models are particularly useful when combined with other exposure assessment method, such as biomonitoring, thus making possible to link exposure concentrations with internal exposure.

1.2.2. Which supplementary tools are available for personal exposure assessment?

Research on human behaviour or activities is a crucial component of modern and future exposure science (Lioy, 2010). The crucial questions are *“Where individuals really are during their daily activities?*”; *“Are concentration peaks of air pollution co-located in time and space with the time period that individuals spend outdoors?”*; “*How much time an individual are exposed in hot-spots?”*. These and other related questions could be answered by the recent development and availability of enhanced resources such as geographic information system (GIS) and global positioning system (GPS), opening thus new insights in the field of personal exposure assessment to air pollution in urban areas.

◢ Geographical Information Systems (GIS)

Geographical Information Systems (GIS) is a useful tool to study the interactions between humans and the environment by providing the required spatial information and analysis. A GIS is an integrated collection of computer software and data used to view and manage information connected with specific locations, analyse spatial relationships, and model spatial processes (Wade and Somer, 2006). All spatial data can be geocoded, i.e. described by x and y coordinates in a geographical coordinate system. In a GIS, different data in databases with geocoded observations can be analysed and visualized. Maps are essential parts in a GIS and can be used as both input and output data.

Air pollution exposure assessment relies heavily on spatial context with the purpose of untangling the associations between air pollution and the individual across space–time. Geographic Information Systems, and associated statistical techniques, along with the availability of spatially referenced health and environmental data, have created unique opportunities to investigate spatial associations between air pollution exposures and health outcomes at multiple spatial scales and resolutions (Collins, 1998; Melnick, 2002). Under the context of exposure research field, GIS allows environmental and epidemiologic data to be stored, analysed, and displayed spatially and temporally, improving data integration and consistency by providing means of capturing and linking spatial data within a single geographical structure. The majority of epidemiological and environmental data has a spatial (location) component, to which GIS adds a powerful graphical and analytic dimension by bringing together the fundamental epidemiological triad of person, time, and the often-neglected place (PHAC, 2008). Also, GIS can be used in combination with dispersion models to simulate the ways in which pollutants propagate in environment, and the exposure as result (Briggs, 2000; Meliker and Sloan, 2011).

Equally, GIS permits spatial linking of different types of data, providing a framework for combining pollution and population data, as required for exposure assessment (Nuckols et al., 2004; Weis et al., 2005; Briggs, 2008; Maantay, 2011; Meliker and Sloan, 2011). GIS allows to create distinct environmental, population and health data layers that can be linked spatially and temporally. Thus, GIS provides the potential to make exposure models more explicitly spatial, and several systems have been developed for modelling exposures in stationary indoor environments (Clench-Aas et al., 1999; Zhan et al., 2006). However, despite its greater applicability, until now, GIS has been used for personal exposure assessment under a place-based perspective, estimating exposure based on geographic proximity between the static location of the individual to pollutant and sources. In such studies, GIS is often used to locate the study population by geocoding addresses (assigning mapping coordinates) (e.g. residence, workplace) and to establish the exposure surrogate on the basis proximity analysis of contaminant source (Jarup, 2004; Weis et al., 2005; Zhan et al., 2006; Hochadel et al., 2006). Several limitations have been identified, including the high aggregation of spatial data, the scale dependence of exposure estimates, the lack of consideration of spatial and temporal variation and the lack of accounting for individual time-activity patterns (Nuckols et al., 2004; Maantay, 2011; Nuvolone et al., 2011).

Recently, GIS have created unique opportunities to derive personal exposure estimates at individual level by offering powerful tools to present spatial information to the level of the individual, conducting predictive modelling, and by integrating information about individuals' time-activity patterns with environmental data. GIS provides access to additional information from a wide variety of sources, such as global positioning systems (GPS) to obtain almost the exact individual´s location at a given time, as discussed below. Some researchers have used GIS with GPS to define time-activity patterns that could feasibly be linked with environmental data for personal exposure assessment (Phillips et al., 2001; Elgethun et al., 2003; Nuckols et al., 2004). Using GIS to spatially integrate individual´s time-activity patterns with environmental data can be helpful in assessing inter and intra-individual variability of exposure to air pollutant in urban areas, reducing uncertainties in exposure estimates, and thus improving the results of epidemiological studies and of risk assessment analyses.

◢ Global Positioning Technology (GPS)

One of the problems of the exposure assessment approaches is the uncertainty related to the human mobility during the exposure assessment period. To overcome this issue, the use of Global Positioning System (GPS) for human tracking presents an enormous opportunity for improving our understanding of how time-activity patterns can influence individual exposure and subsequent health effects. GPS is a freely accessible and promising technology by monitoring individual´s real-time geographic positions. This technology uses differences in timing data of radio signals that are transmitted from a constellation of satellites to determine an individual's location. As technology progresses, a GPS receiver/data logger can integrated into watch, wear or mobile phone (USEPA, 2003).

Predictability in human dynamics by studying the mobility patterns of individuals using GPS equipped mobile phones became an emerging field (Gonzalez et al., 2008; Song et al., 2010). GPS-equipped mobile phones can record the latitude-longitude position of individuals at each moment, offering many advantages over traditional time-location analysis, such as high temporal resolution, and minimum reporting burden for participants (Rainham et al., 2010; Chaix et al., 2013). This information can be logged passively or sent in real-time using cell phone networks to a remote server for further analysis, and allows researchers to map an individual's space-time path through multiple contexts.

Collection of time-location information using GPS technology provides continuous tracking of the individuals with high data resolution in time and in space. The GPS technology guarantees that there will be an increasing availability of large amounts of data affecting to individual trajectories, at increasing localization precision. However, it has be emphasized that a GPS is not a standalone toot to determine time-activity locations, such a commuting, indoor or outdoor locations, since it can only give information on the path that a moving individual follows through space as a function of time, i.e. GPS trajectory (Wu et al., 2010; Rainham et al., 2010; Zheng and Zhou, 2011). Significant uncertainties associated with the processing and classifying of GPS trajectories is one of challenging issue for the exposure studies (Wu et al., 2010).

Recently, GPS technology has been used successfully in personal exposure assessment to collect individuals' time-location information (Amorim et al., 2012; Valente et al., 2012). Several personal exposure studies have used a well-designed integration of GPS devices with portable pollutant monitors to determine potential exposure at the individual level (Greaves et al., 2008; Boogaard et al., 2009; Lioy, 2010; Dons et al., 2011; Zwack et al., 2011; Broich et al., 2012; Cole-Hunter et al., 2012; Miranda et al., 2012). The development of portable personal exposure monitoring devices is a fast evolving field and incorporates everyday devices, such as smartphones. An example is the portable, real-time exposure monitoring system which was developed and described by Negi et al. (2011). This device communicates wirelessly with a smart phone which serves as user interface as well as for processing monitoring data, adding GPS information and to display concentration profiles (Negi et al., 2011).

Overall, combined with GIS, GPS technology are expanding their applications as supplementary tools for personal exposure assessment emerging as model input for personal exposure studies based on individual movement patterns or routes, as detailed discussed in Section 1.3.2. Despite some limitations of GPS technology, findings show that personal exposure profiles towards changing environmental influences, which differ from other individuals as well as the population average, can be derived by using a GPS approach, and suggest that GPS can be seen as the way forward (Dons et al., 2011; Richardson et al., 2013).

Summary 1.2.: *Personal exposure estimation is a crucial component to quantify exposure-related health effects. A new context of exposure assessment recognizing importance of the actual spatial and temporal scales on quantifying personal exposure to air pollution is emerging. Currently, personal exposure assessment methods can be aggregated in two main groups: dynamic personal* *exposure approach and static personal exposure approach. On a static approach, time-activity patterns of individuals are not directly addressed, while dynamic approach takes into account individual activities in space and time and, therefore explicitly addresses spatio-temporal variations in esposure. The availability of supplementary tools for personal exposure assessment such as geographic information system (GIS) and global positioning system (GPS) enhances the characterization of variable air pollution levels and individual's time-activity patterns, as required by personal exposure assessment. Combining GPS with GIS offers the opportunity to take a step forward in the quantification of personal exposure to air pollution in urban areas.*

1.3. Modelling: a priority area for personal exposure research

Modelling is a very important tool in exposure and health impact assessment research since it is a flexible and cost-efficient indirect method for assessing human exposure. An exposure model is "*a logical or empirical construct which allows estimation of individual or population exposure parameters from available input data*" (WHO, 2000). Technological advancements in computing processing power, availability of human activity/environmental data have allowed the development and application of comprehensive exposure modelling system to provide both spatially and temporally resolved exposures. Human exposure modelling is presented thus as promising tool to address the high temporal and spatial variability in the personal exposure imposed by the urban environment and has become a fundamental and required approach of exposure analysis as it provides an efficient and economical means for assessing exposure of individuals over a variety of spatial and temporal scales for past, current, future, or hypothetical conditions.

Personal exposure modelling allows quantifying how much atmospheric air is contaminated in different locations of the study area, and simulating how different individuals interact with those air pollution levels to derive personal estimates of its exposure during the study (USEPA, 2004b). Exposure modelling is typically used to supplement personal or biological monitoring data or when such measurements are not available/appropriate for the exposure assessment situation. Thus, exposure models are essential for comprehensive exposure assessment because we will never be able to monitor or measure every exposure everywhere. Additionally, they also play an essential role in establishing guidelines for acceptable levels of indoor and outdoor air pollution, which rely on the estimation of health risk associated with air pollutants for different possible scenarios.

The crucial purpose of personal exposure modelling is to reflect “real-world” human exposure to air pollutants over time and consequently assess the health outcomes of air pollution exposure (Nethery et al., 2008). Thus, the need for personal exposure models increases proportionally with the growing knowledge of the importance of the spatial and temporal scales imposed for a variety of indoor and outdoor environments and time-activity patterns for personal exposure assessment. Generally, to address these challenges, exposure models incorporate one or more of the three fundamental variables that govern human exposure: (i) pollutant source identification and emission rate, (ii) outdoor and/or indoor pollutant concentrations, and currently (iii) human activity, as presented in Figure 1.9.

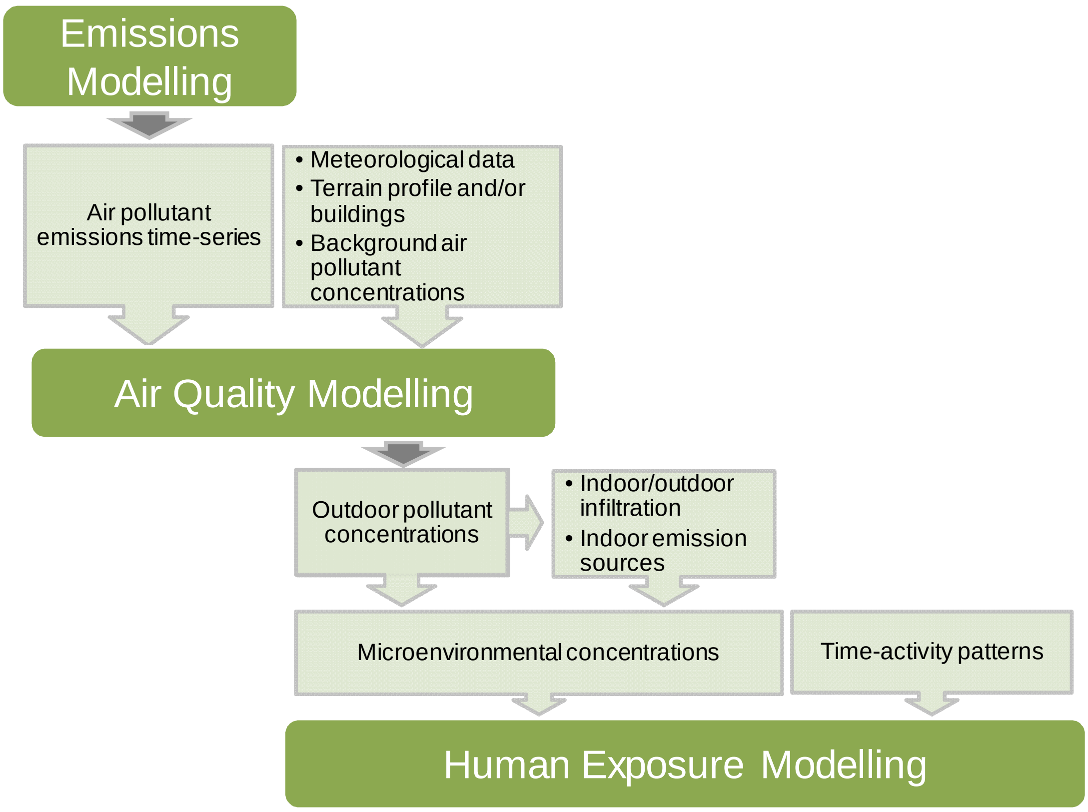

Figure 1.9. Link between the principal components of an exposure model.

1.3.1. Air Quality Modelling: How it may contribute to personal exposure assessment?

Air quality modelling allows establishing the relationships between current emissions and current air quality at particular locations. Air quality models serve multiple purposes in exploring the relationships between air pollutants and exposure-related health effects. One important application is extending observations spatially to reduce exposure errors and uncertainties that arise from the limited spatial coverage of current routine monitoring networks. Further, air quality models play a key role in identifying the most efficient and cost-effective strategies for reducing source emissions and protecting human health and welfare, thus serving as an important management tool (USEPA, 2009a).

Air quality models describing the dispersion and transport of air pollutants in the atmosphere can be distinguished on many grounds: on the spatial scale (local, urban, mesoscale, regional, global); on the temporal scale (episodic models, (statistical) long-term models); on the treatment of various processes (chemistry, wet and dry deposition); and on the complexity of the approach used for the physical process description. Depending on the modelling objectives, it is important to select an appropriate model from among the considerable diversity of the available tools taking into account the simplifications and assumptions considered by the model (Borrego et al., 2001).

In general term, air quality models can be divided into (i) process oriented models and (ii) statistical models (EEA, 1996; Daly and Zannetti, 2007; Solomon et al., 2012). Process oriented models are based on the description of physical/chemical processes: starting with emissions, atmospheric advection and dispersion, chemical transformation and deposition are calculated. This type of models is able to give a description of cause-effect relations. Statistical models are valuable tools in diagnose of air quality by means of interpolation and extrapolation of measuring data (e.g. the concentrations measured show a statistically significant dependence on the volume of traffic). Each of these modelling approaches has been used to characterize air quality concentrations for personal exposure modelling to air pollution.

A statistical model may be applied to time-series obtained from measurements for the purpose of establishing a relationship among dependent and independent variables. It is both a strength and a weakness of statistical models that they do not require nor imply any causal relationships between the model variables. Statistical models require both input and output variables to be known in the model development system. However, this type of models should be used with caution. They may be considered valid only within the range of the data from which they were derived. That is, the interpolation between data values is acceptable, but extrapolation to a set of conditions outside the range of data may yield invalid results. Interpolation models and land use regression models are examples of a statistical modelling.

Interpolation models utilize measurements at multiple locations throughout the study area and estimate pollutant concentrations for unmeasured locations (Briggs, 2000). Estimations are derived from spatial trends within the measured data. There are many spatial interpolation methods including the local neighbourhood approaches (e.g. inverse distance weighting), the geostatistical approaches (e.g. kriging), and the variational approaches (e.g. thin plate spline) (Coulibaly and Becker, 2007). Several studies have predicted estimates of personal exposure using spatial interpolation of air quality data (Finkelstein et al., 2003; Künzli et al., 2005; Jerrett et al. 2005b; Cohen et al.,2009; Son et al., 2010), but there is not yet consensus on which methods are most appropriate. Moreover, the quality of estimated concentrations is related to the degree of monitor coverage and spatial heterogeneity of the pollutant within the study area (Wong et al., 2004; Son et al., 2010).

Land-use regression (LUR) is an empirical modelling approach being used to address the limited spatial coverage found in routine air quality monitoring networks. This approach uses auxiliary data on a city's physical characteristics to estimate pollutant levels in relation to local activities (Crouse et al., 2009). These models spatially link ambient pollutant concentration measurements throughout the study area with other associated variables such as distance to pollutant source, topography, building types, population density, socio-economic status, land use, traffic volume within GIS (Brauer et al., 2007; Ryan and LeMasters, 2007; Hoek et al., 2008a). Recent applications have incorporated physically based factors such as meteorology and topography in an attempt to improve estimates (Arain et al., 2007; Ryan et al. 2008). LUR models treat the pollutant of interest as the dependent variable and proximate land-use, traffic, and physical environmental variables as independent predictors. As a result, they predict pollution concentrations at a given site based on surrounding land use and traffic characteristics (Jerrett et al., 2005a; HEI, 2010). Applications have demonstrated a good agreement between measured and modelled benzene and organic compounds, although NO_2 is more challenging (Crouse et al., 2009). However, there are several limitations to this type of models. Namely, even though LUR models offer improved spatial resolution, they still may not capture a small enough spatial scale for individual exposure assessment (Brauer et al., 2007; Hoek et al., 2008b).

Process oriented models include the traditional air dispersion models, and use the best available emission estimates and local meteorological data to predict pollutant concentrations at various locations. Over statistical models, air dispersion models have the main advantage to incorporate both spatial and temporal variation of pollutant concentrations and can be used to assess time periods from hourly averages to annual periods. Air dispersion models are one of the most common types of models used for air quality management and have been established as the primary method for assessing human exposure in urban areas (Kousa et al., 2002; Jerrett et al., 2005a; Zou et al., 2009a). Air dispersion models estimate pollutant concentration profiles over space and time by applying mathematical equations based on physical processes to site specific input data.

Air dispersion models can generally be categorised by their type (e.g. Gaussian, Lagrangian, Eulerian) and scales of application (Denby et al., 2011). Gaussian model is one of the mostly used air quality model based on the process oriented approach. They assume that the concentrations from a continuously emitting source are proportional to the emission rate, inversely proportional to the wind speed, and that the time averaged pollutant concentrations horizontally and vertically are well described by Gaussian distributions (Boubel et al.,1994; Nieuwenhuijsen, 2003). In its simplest form, the Gaussian plume model assumes that there are no chemical or removal processes taking place and that pollutant material reaching the ground or the top of the mixing layer as the plume grows is reflected back towards the plume centreline. Gaussian models are more suitable for calculating annual mean concentrations in an urban region than for calculating of hourly mean concentrations. The ADMS-URBAN (European model) (McHugh et al. 1997) and AERMOD model (recommended by the USEPA) are examples of Gaussian models.

The Eulerian and Lagrangian approaches are more physically realistic, but numerically complicated and computationally expensive (Figure 1.10) (Seinfeld and Pandis, 2006). Eulerian and Lagrangian models can provide realistic simulations of the atmospheric transport and mixing of air pollutants at several scales (Borrego et al., 2006). In an Eulerian model, chemical species are transported in a fixed frame of reference, usually the surface of earth (Figure 1.10). This enables easy representation of the pollutant production and transformation processes. The space domain (geographical area or air volume) is divided into "small" squares (two-dimensional) or volumes (three-dimensional), i.e. grid cells. Most Eulerian models use a grid system to describe atmospheric dynamics (advection and diffusion), emission sources and chemical production, and generate four-dimensional (space and time) trace species concentrations fields for each of the species modelled (Seinfeld and Pandis, 2006). These models use numerical terms to solve the atmospheric diffusion equation (i.e. the equation for conservation of mass of the pollutant) (Seinfeld and Pandis, 2006). The numerical solution of the transport term in the Eulerian framework becomes more difficult and often requires substantial computational resources to be accurate enough compared to the Lagrangian approach. The main advantage of the Eulerian models is the well-defined three dimensional formulations which are needed for the more complex regional scale air pollution problems. Long range transport simulations are mostly done using Eulerian models. Example of Eulerian models are the TAPM (Hurley, 2008), CAMx (Ferreira et al., 2012) and CHIMERE model (Monteiro et al., 2007).

In Lagrangian models, also called Lagrangian Particles or Random Walk model, the motion of air masses or particles following the flow is studied (Figure 1.10). In these models, the concentration is computed by counting “fictions particles” (computer-particles) in a user defined volume (e.g. the cell of a regular grid). Each “particle” represents a particular mass of one or several pollutants emitted from a given source. Hence, transport caused by both the average wind and the turbulent terms due to wind fluctuations is taken into account. Time-dependent trajectories of particles are computed by stochastic differential equations (Langevin equations), which aim at describing turbulence properties (Degrazia, 2005).

The computation time in Lagrangian models is directly linked to the number of particles within the model domain, which in turn is determined by the number of particles released, the size of the model domain and the wind speed. This type of models should provide a better description of the dispersion and transport of pollutants than the simpler Gaussian models, particularly in complex terrain (Degrazia, 2005; Daly and Zannetti, 2007). Also, to determine pollutant concentrations in street canyons or urban blocks, high resolution flow models that can resolve buildings need to be applied (e.g. computational fluid dynamics (CFD) models) (Borrego et al., 2003; 2004; Martins et al., 2009). This type of models is particularly useful for simulating short-term releases from sources with highly variable emission rates in complex dispersion scenarios (Degrazia, 2005). Moreover, these models begin to be used for regulatory purposes in some European countries such as the Official reference model of the German Regulation on Air Quality Control, the AUSTAL2000 model (Janicke and Janicke, 2002; Janicke, 2004). Also in this study, AUSTAL2000 was selected to simulate the air pollution dispersion. Despite the high computational requirements for this model, its applicability to simulate the air pollution dispersion in areas with complex topography, its high flexibility in modelling the physical processes involved, as well the fast processing of the input data (e.g. buildings and emission sources characterization), were decisive for the choice of this model in pursuit of the objectives set in this research.

Overall, air dispersion models offer improved spatial and temporal resolution to estimate air pollutant concentrations in locations without dense monitoring networks (Clench-Aas et al., 1999; HEI, 2010). From a comparative evaluation of the performances of four methods for exposure assessment of air pollution, Zou (2010) shows that air dispersion models provide the most reliable exposure impact simulation results, and its accurate performance was attributed to data input requirement. Therefore, air dispersion modelling presents a promising tool to personal exposure assessment by characterizing the air pollution levels required to quantify exposure at the individual level and by helping to identify high exposure scenarios (i.e. high exposure sites, meteorological conditions that lead to high pollutant concentrations), as well as to provide high-resolution analysis of patterns in health outcomes and environmental factors (Hruba et al., 2001; Lipfert et al., 2006; 2008).

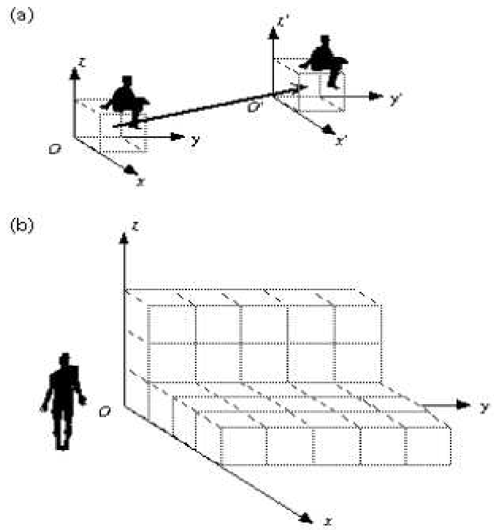

Figure 1.10. a) In the Lagrangian system the observer follows movement of air parcel, and b) in the Eulerian system, the observer studies atmospheric motion at a fixed reference point (Seinfeld and Pandis, 2006).

1.3.2. Personal Exposure Modelling: From a place to individual-based approach

Over the past 20 years, several exposure modelling methods have been developed with the aim of estimating exposure at the individual level. The major purpose of these models is to characterize air quality concentrations to be used as surrogate of personal exposure to air pollution and assumes that subjects within a demographic area (e.g. census units) are equally exposed to air pollution. Thus, over the past decade, air quality models have been integrated with GIS in attempt to reflect individual exposure by combining air pollutants concentration data with residence location (e.g. Bartonova et al., 1999; Gauderman et al., 2007; Hoek et al., 2008a). Nevertheless, the knowledge of where individuals spend time is essential for the assessment of human exposure to air pollution and research on human behaviour or activities is a crucial component of modern and future exposure science (Lioy, 2010). Thus, individual-based personal exposure modelling, although data and computer intensive, is considered the closest to a "best" estimate of personal exposure to air pollution (Jerrett et al., 2005b; Özkaynak et al., 2008; HEI, 2010).

To mitigate the problem of a place-based exposure approach, the concept of microenvironment was developed (Georgopoulos and Lioy, 1994; Valente, 2010). An individual's daily activities are related to a series of microenvironments, such as home, workplace, in vehicle during travelling route, and recreation place. Microenvironments are defined as a location where the concentration of an air pollutant is considered to be spatially homogenous during the time that individuals are exposed (Kaur et al., 2007; Edwards et al., 2001; 2005). Despite the fact that time-activity location analyses are very complicated, microenvironment approach use microenvironments, typically indoor residences, indoor workplaces, other indoor locations, outdoor near residences, other outdoor locations, and in vehicles, as a proxy of time-activity patterns (Srivastava, 2005; Zou et al., 2009b). Under the microenvironment assumption, individual´s air pollution exposure is calculated using a similar approach as represented by Equation 1.1 but considering the discrete product of “representative” concentrations for the individual or activity being examined in that microenvironment times the duration of the time spent there (Hertel et al., 2001; Weisel, 2002):

$$E_i = \sum_{j=1}^{m} c_{ij} t_{ij} \tag{1.3}$$

where E_i (units: ppmh or µg.m^{-3}·h) is the personal exposure for person i over the specified period of time, c_{ij} is the air pollution concentration (units: ppm or µg.m^{-3}) in each microenvironment j, t_{ij} is the time spent (units: h) by person i in each microenvironment j, and m is the number of different microenvironments.

Several individual-based personal exposure models based on a microenvironment approach, including AirPex (Freijer et al., 1998), SHEDS-PM (Burke et al. 2001), HAPEM (Özkaynak et al., 2008), APEX (USEPA, 2009b), are available. These models are designed to simulate the distribution of personal exposure in several microenvironments (e.g. outdoors, traffic environments, indoor-residential, public buildings, workplaces, and schools) (Burke et al., 2001), by combining the time spent at visited microenvironments and the estimated pollutant concentrations (e.g. PM10, VOCs, etc.) at every microenvironment. Usually, microenvironmental concentrations are estimated as a combination of infiltrated outdoor air and indoor source emissions based on mass balance or empirical indoor/outdoor relationships. Additionally, the time spent at visited microenvironments and activities of individuals used by these approaches is obtained based on time–activity databases (e.g. Consolidated Human Activity Database (CHAD); National Human Activity Pattern Survey (NHAPS)) (Burke et al., 2001; Kruize et al., 2003; Klepeis, 2006; Özkaynak et al., 2008). However, by using this time-activity location data, individual air pollution exposure context can be assumed as a series of independent microenvironment exposures (Ballesta et al., 2008).

Also, Zidek et al. (2005) presents a stochastic approach for estimating personal exposure, the pCNEM model, based on time–activity databases (i.e. NHAPS). In this model, microenvironmental concentrations are estimated using a mass-balance indoor model and the closest measurement station as proxy for the outdoor concentration. The individual´s location is addressed by distinguishing between home and workplace and identifying the districts that are associated to the nearest pollution monitor. This model enables the estimation of personal exposure for randomly picked individuals by running the stochastic model several times on similar diaries of the same population subgroup. However, individuals who belong to same population subgroup may have different time-activity patterns and, consequently different air pollution exposure levels (Kwan, 2009). Also, the aim of their model is to give probabilistic estimates for certain population subgroups instead of modelling time and space variant exposure dynamics of a specific individual person.

The advent of GIS provides the potential to make these models more explicitly spatial. As a first attempt to model individual exposure on a very detailed spatio-temporal resolution, the spatio-temporal exposure model system (STEMS) (Gulliver and Briggs, 2005; 2011), was developed. The STEMS model is a GIS-based system that simulates the exposure of an individual or subpopulation to traffic-related pollution as people travel through a dynamic pollutant field. STEMS incorporates an air dispersion model (ADMSUrban), an empirical background pollutant model (BACKGAMON), traffic model (SATURN) (a model for vehicle flows), and a time-activity model (TOTEM). Time–activity patterns are simulated for individuals over an appropriate period (e.g. week, day, or part day), based on results from time–activity surveys. Exposures are then estimated for each location by cross-reference to the pollution map for that time period. Although the modelling approach had great promise, the current version of this model only focuses on journey-time exposure to PM10 (i.e. during on foot or in a vehicle), and 24-h exposure profiles are not provided. Also, indoor sources are not considered for personal exposure assessment, estimating indoor concentrations (i.e. in vehicles) by using outdoor concentrations and weighting factors.

Recently, there has been an increasing focus on using GPS technology to collect the individual trajectory information to be used in combination with air pollution levels to estimate personal air pollution exposure levels in urban areas. A traffic air pollution exposure modelling system named AirGIS was developed by National Environmental Research Institute in Denmark (Jensen, 2006). AirGIS system included two modules. One to simulate urban air pollution levels using Danish Operational Street Pollution Model (OSPM), road network, traffic information, and a Geographic Information System (GIS). The second module estimates personal exposure at address level (including about 200,000 addresses in Denmark) and with one hour of time resolution. Also, apart from modelling exposure at address level, the system includes a model system for the estimation of exposure under transport along a route provided by individual carried cell phones with built-in GPS receivers, which send location information by short message service (SMSs) to AirGIS tracking centre at twenty seconds intervals. Despite of AirGIS project is promising to collect individual-based real-time positioning, this system can only be applicable in Denmark and for smaller field studies (Hertel et al., 2008). The personal exposure in stationary microenvironments is estimated under a place-based approach, considering the location address and only for home and workplace. Also, SMSs with individual trajectory data are only sent when the subject is moving out of a defined area and has associated costs as positions. Also, only vehicles with GPS technology can be considered for exposure analysis.

Gerharz et al. (2009; 2013) developed an initial framework for spatio-temporal individual exposure modelling, taking GPS data and information from TADs and questionnaires, indoor, and outdoor concentration into account. For the outdoor distribution, a dispersion model was used and extended by actual ambient fixed site measurements. Indoor concentrations were modelled using a simple mass balance model with the estimated outdoor concentration fraction infiltrated and indoor activities estimated from questionnaires. Information on time-activity patterns was provided from a combination of GPS data and self-administered TADs. The entries of the diaries are classified into visited activities relevant for the exposure model, distinguish home, working environment, other indoor, transportation, and outdoor. This information is posterior used to identified indoor environments in GPS processed data.

Daily average exposure values estimated by Gerharz et al. (2009) evidence a strong influence of individual behaviour. However, there are limitations to the general applicability of this methodology due to simplifications and assumptions adopted such as the qualification of indoor activities for which the TAD was used and where the GPS sensor cannot receive a signal. This model is strongly dependent on TADs and questionaries' information to derive individual activity profile, providing exposure estimates only if the individual resides in a microenvironment which is specified in the model (Gerharz et al., 2013). Also, although GPS trajectories are analysed and processed (Wu et al., 2010), the microenvironments are identified based on information provided by TADs, which has several weaknesses (Section 1.1.3.) and only indoor and in-vehicle microenvironments are identified, ignoring exposure during walking periods.

Summary 1.3.: *To assess and manage health effects associated with current and emerging complex air quality issues, personal exposure modelling has become a priority and required approach of exposure analysis, as it provides an efficient means for assessing personal exposure at the spatial and temporal scale imposed for a variety of "microenvironments" during individual´s time-activity patterns. In this context, air dispersion modelling play a key contribution to personal exposure assessment in order to characterize the air pollution levels required to quantify exposure at the individual level. Several personal exposure models have been developed, presenting crucial strengths over other personal exposure methods. Clearly, personal exposure modelling has progressed significantly over the past decades, from crude qualitative estimates to today´s refined integrated methods yielding more accurate quantitative exposure estimates at the individual level. Instead of a place-based personal exposure approach, individual-based exposure models consider time-activity patterns of the individual to obtain more realistic spatio-temporal individual exposure estimates. Recent information technologies, namely GPS, facilitate the collection of individual´s spatio-temporal trajectory, and when combined with air pollution levels can effectively derive individual-level personal exposures. However, until now several efforts on characterizing the spatial and temporal distributions of air pollution have been expended, but much work remains in understanding the role of individual mobility in conditioning exposures in urban areas. Also, very little has been done toward validating of such models at the level of the individual. The validation of models with independent data sets is useful to check whether the proposed models serve as surrogates for individual exposure and to know the extent of the exposure estimation error, which should be accounted for in health impact assessment. Under this framework, accurately quantifying human exposure to air pollution in urban areas still remains a challenging task. Consequently, the development of personal exposure models that provide a better understanding of exposure by establishing source-receptor relationship and by explicitly preserving the sequence of exposure events at the individual exposure level in the urban environment is a priority area for future exposure research.*

1.4. Research Objectives and Thesis structure

The prime objective of this research work is the development of a consistent approach for the quantification of individual exposure to traffic-related hazardous air pollutants in urban areas within distinct microenvironments by using a novel methodology for trajectory analysis of the individuals in order to support health impact assessment and decision-making in public health management.

To achieve the defined objective, the following tasks were accomplished:

- An overview of the currently available methodologies for the quantification of personal exposure to air pollution. At this stage, the research was focused on different personal exposure methods and supplementary tools available. The dynamic exposure approach was evaluated in comparison with static exposure methods;
- Identification of the relevant parameters of exposure quantification at urban scale, such as spatial and temporal resolution of the data. The final use of the results, including health impact assessment requirements, was considered for this purpose;
- A comprehensive analysis and identification of the current and future potential impacts on human health associated with exposure to air pollution. This analysis was based on an atmospheric and health impact assessment modelling contributing to a better understanding of the number of deaths that are attributable to the exposure to current air pollution levels and under future climate in Portugal;
- Development and implementation of a new module into the Transport Emission Model for Line Sources (TREM) to quantify emissions of traffic-related hazardous air pollutants (HAPs), providing detailed information on HAPs emissions with higher resolution within urban areas;
- Development of a new personal exposure modelling tool based on trajectory analysis of individuals and air pollution modelling with high spatial-temporal resolution to provide the magnitude, frequency and the intra and inter-variability of individuals' exposure levels that is essential for health impact assessment. The development and implementation of trajectory data mining and geo-spatial analysis algorithm within Geographic information system was performed at this stage of the research, in order to process the trajectories obtained with Global Positioning System and collected by mobile-phones;
- Characterization of the variability of the microenvironmental parameters based on a probabilistic approach providing an additional knowledge on the variation associated with microenvironmental concentrations and its contribution to the individual exposure estimates;

- Application of the exposure model to the study area. Based on the data provided by the transportation – emission – air dispersion modelling and the daily trajectories of the individuals, statistics on individual's air pollution exposure were estimated for each individual;

- Validation of the developed exposure modelling tool by using personal and biological exposure measurements collected during the daily activities of individuals in a measurements campaign. The exposure modelling tool presents as a useful tool to be used in combination with personal monitoring and biomonitoring, enabling to analyse and understand the exposure measurements obtained.

This study is presented in seven distinct chapters, based on published and submitted manuscripts.

A comprehensive analysis of the current impacts on human health associated with exposure to urban air pollution is performed in Chapter 2. Thus, a health impact assessment is conducted in Chapter 2 in order to quantify the potential health benefits by meeting the air quality limit values (2008/50/CE) for short-term PM10 exposure in an urban area. Additionally, in order to identify the relevant parameters of exposure quantification at urban scale, the role of the population mobility and inhomogeneity of spatial pollution pattern is analysed and considered in health impact assessment. The air pollution spatial variation and high population mobility observed within urban areas are identified as important factors for the short-term health risk analysis. Therefore, an improved methodology to process the population data taking into account daily average population mobility and to process air quality time series to obtain representative background pollution values are presented in Chapter 2. The main outcomes of this chapter highlight the importance to study the human mobility and inhomogeneity of spatial pollution pattern to improve estimations of human exposure to air pollution in urban areas, thus providing relevant information for the research performed in the next chapters.

The identification of the future potential health risk under climate-induced changes in air pollution levels within urban areas are analysed and discussed in Chapter 3. This analysis was based on an atmospheric and health impact assessment modelling conducted to understand the potential impacts of climate-induced changes in PM10 concentrations and how future changes in PM10 concentrations contribute to mortality attributable to urban air pollution in future scenarios. Worldwide, several studies have already discussed the relationship between the climate change and health effects. However, studies focusing on the health impacts of air quality in Portugal are very few. Thus, this chapter intends to contribute to a better understanding on the number of deaths that are attributable to the exposure of air pollution levels under future climate in Portugal, emphasizing the importance of indirect effects of climate change on human health.

A fundamental question addressed in Chapter 4 is to what extent urban air pollution is affected by road traffic sources. In this concern, the characterization of the transport activity and the quantification of corresponding emissions in urban areas where inhabitants are leaving close to the pollution sources combined with air quality modelling allows establishing the relationships between current emissions and current air quality at particular locations, which is crucial for human exposure analysis to traffic-related air pollution in urban areas. In this scope, and given the known toxic and carcinogenic effects of HAPs on human health, Chapter 4 is focused on the development of a modelling approach to quantify emissions of traffic-related hazardous air pollutants in urban areas considering complex road network and detailed data on transport activity. A new version of the Transport Emission Model for line sources has been developed for hazardous pollutants (TREM-HAP). Also, this new version of the model was extended to integrate a probabilistic approach for the uncertainty quantification using Monte-Carlo technique. Thus, a probable distribution of the emissions of benzene, 1,3-butadiene, formaldehyde, acetaldehyde, acrolein, naphthalene and also particulate matter (PM2.5) for different types of roads considering vehicle technology mix, driving conditions and traffic volume fluctuations is presented in Chapter 4. In addition, the important contribution of cold start emissions to the total daily values of HAPs is investigated.

Once recognized the spatial and temporal scales required by the exposure events, a new exposure modelling tool, the GPS based Exposure Model to Traffic-related Air Pollution model (ExPOSITION) are developed and discussed in Chapter 5 in order to quantify the short and long-term exposure to traffic-related air pollutants at the temporal and spatial scale imposed by the individual. Hence, the Chapter 5 presents the development and application of a new modelling tool for quantification of human exposure to traffic-related air pollutants within distinct microenvironments by using a novel approach based on trajectory analysis of individuals and air pollution modelling with high spatial-temporal resolution. For this purpose, information on pollutant concentrations at different microenvironments and detailed time-location data collected for each individual by mobile phones with Global Positioning System technology are processed using trajectory data mining and geo-spatial analysis within Geographical Information System to obtain time-activity patterns. The detailed emission data provided by the TREM-HAP emission model are considered as important inputs to AUSTAL2000 dispersion model to provide information on variability of outdoor air pollutant concentrations. Additionally to outdoor, pollutant concentrations in distinct indoor microenvironments are characterised using a probabilistic approach to estimate the variability of the microenvironmental parameters in the predicted individual exposure.

To evaluate the feasibility of the developed exposure model, Chapter 6 includes the application and validation of the new exposure modelling approach for benzene, which is defined as one of the most important health-based European Union priority substances, against personal exposure measurements and biological monitoring data collected during the daily activities of individuals in a measurements campaign. In addition to road transport emissions, vehicle refuelling emissions were also considered in the current research in order to guarantee completeness of the benzene emission estimations. The modelling cascade, including transportation-emission-dispersion-exposure models are applied to a selected urban area in Portugal.

Finally, in Chapter 7 a brief summary of the main results is presented. Additionally, the general conclusions are explored and possible future developments discussed.

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CHAPTER TWO

2. QUANTIFICATION OF HEALTH BENEFITS RELATED WITH REDUCTION OF ATMOSPHERIC PM10 LEVELS: IMPLEMENTATION OF A POPULATION MOBILITY APPROACH

Published

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Abstract

This study is focused on the assessment of potential health benefits by meeting the air quality limit values (2008/50/CE) for short-term PM10 exposure. For this purpose, the methodology of the WHO for Health Impact Assessment and APHEIS guidelines for data collection were applied to Porto Metropolitan Area, Portugal. Additionally, an improved methodology using population mobility data is proposed in this work to analyse number of persons exposed. In order to obtain representative background concentrations, an innovative approach to process air quality time series was implemented. The results provide the number of attributable cases prevented annually by reducing PM10 concentration. An intercomparison of two approaches to process input data for the health risk analysis provides information on sensitivity of the applied methodology. The findings highlight the importance of taking into account spatial variability of the air pollution levels and population mobility in the health impact assessment.

Keywords: air pollution; health impact assessment; mortality; particulate matter; population mobility, background concentrations.

2.1. Introduction

Over the last few decades, human exposure to particulate air pollution has been identified as a risk factor for human mortality and morbidity, as well as broad range of negative health outcomes at levels usually experienced by urban populations due to short and long-term exposure to particulate matter was established (Künzli et al., 2000; Anderson et al. 2004; 2005; Pope and Dockery, 2006; Samoli et al., 2008). The recently adopted European directive (2008/50/CE) revised the limit values for PM10 previously defined by Framework Directive (1999/30/EC) and set up new quantitative standards for PM2.5. Nevertheless, PM thresholds levels to which exposure does not lead to adverse effects on human health have not yet been identified and given that there is a substantial inter-individual variability in exposure and in the response, it is unlikely that any standard or guideline value will lead to a complete protection for every individual against all possible adverse health effects of particulate matter (WHO, 2006).

A few recent studies have reported a strong epidemiologic evidence of a causal link between particulate air pollution and mortality (Boldo et al., 2006; Jusot et al., 2006; Dockery, 2009), thus providing quantitative estimates of the health effects related to air pollution. The Air Pollution and Health: A European Information System (APHEIS) project showed that 1150 premature deaths could be prevented annually considering a cumulative short-term exposure if daily average PM10 concentrations in the 23 European cities will be reduced to 50 $\mu g.m^{-3}$. The long-term impact would be even higher, totalling 21 828 of premature deaths prevented per year if annual mean PM10 concentration will be reduced to 20 $\mu g.m^{-3}$ (APHEIS, 2005). However, no Portuguese cities were included in the European study and only little information concerning the impact of environmental factors on human health has been published for Portugal (Alves and Ferraz, 2005; Nogueira et al., 2005; Casimiro et al., 2006; Trigo et al., 2009; Alves et al., 2010).

To estimate the health impact of atmospheric pollution on population, the prior knowledge of different variables, such as exposure concentrations time series, number of people exposed, current mortality rates for each health indicator and quantitative estimates for the association between the exposure and health effects are required. Additionally, it is important to determine the relationship between the exposure concentration, which vary substantially with geographical location, and the exposure duration which is related with human activities. Therefore, population mobility is one of the factors that may affect significantly the exposure and should be considered in risk assessment (Boudet et al., 2001; Jerrett et al., 2005a; 2005b; 2005c; Krewski et al., 2005).

The present study provides a quantitative assessment of potential health benefits related with the reduction of short-term exposure to inhalable particles (PM10) in Porto Metropolitan Area (Portuguese: Área Metropolitana do Porto, or AMP). For this purpose, WHO methodology for quantitative assessment of the health impact related with air pollution was applied to the study area. The input information was processed in accordance with Apheis guidelines for data collection (Medina et al., 2001). Additionally, an alternative approach to process the population data taking into account daily average population mobility and an innovative approach to process air quality time series to obtain representative background pollution values have been proposed in this work in order to improve estimations of population exposure.

2.2. Methodology

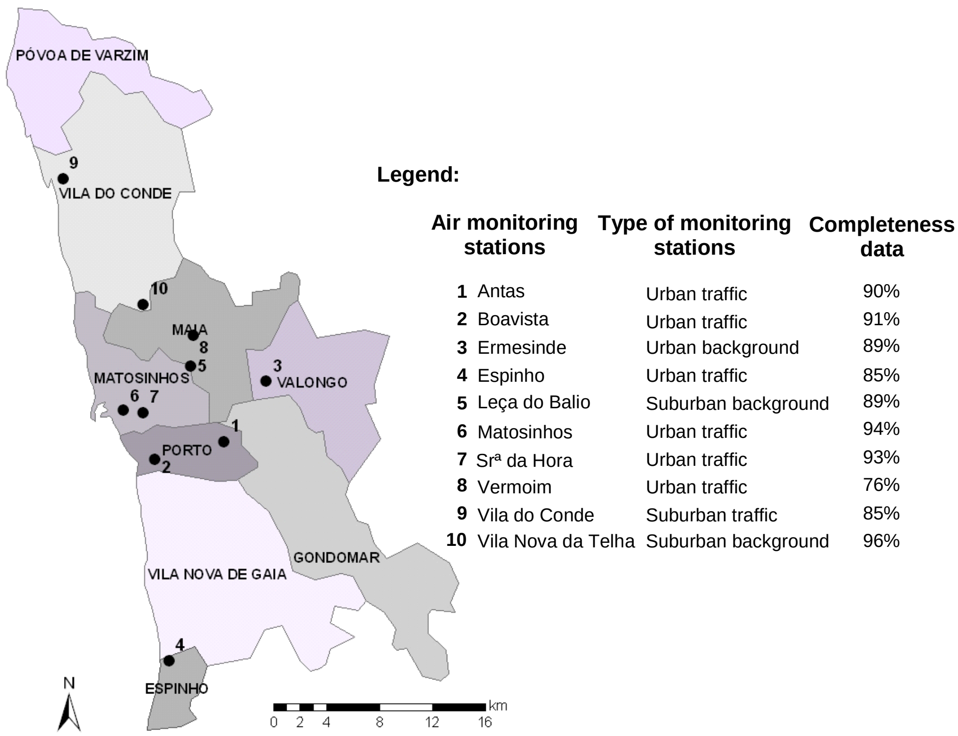
The Porto Metropolitan Area was selected in this study for the heath impact assessment. It is the second largest population agglomeration in Portugal and is characterised by frequent occasions of daily PM10 levels exceeding the limits as defined by Directive 2008/50/CE. Because of the data availability, the study period is focused on 2004. At this period, AMP was constituted by the nine municipalities with a total area of 814.5 km^2 (Figure 2.1). The resident population of AMP in 2004 was about 1,272,176 thus comprising about 10% of the national population.

2.2.1. Quantification of attributable cases prevented

A methodology to quantify health effects is conducted in terms of number of cases attributable to air pollution that may be prevented by reducing current levels of PM10 (Künzli et al., 2000; APHEIS, 2005). An estimate of attributable deaths (AD) is obtained from the average number of deaths ($\bar{y}$), the regression coefficient β provided by epidemiological studies that characterise the ratio for a unit increase in pollutant concentration, and the difference between the daily average concentration ($\bar{x}$) and a reference value under given scenario (x^*):

$$\mathrm{AD} = \bar{y} \times \beta\left(\bar{x} - x^*\right) \tag{2.1}$$

The EIS-PA model, developed by French Surveillance System on Air Pollution and Health as a support tool for automated and standardized health risk assessment (INVS, 2000), is used in this study to calculate the number of premature deaths prevented annually due to the reduction of PM to the selected “target” concentration. The results of EIS-PA model application provide estimates of the health outcomes related with short-term (1-2 days) and cumulative short-term (40 days) exposure. The input data on air quality, population and mortality rates used for the modelling are described in the following sections.



Air monitoring stations	Type of monitoring stations	Completeness data
1 Antas	Urban traffic	90%
2 Boavista	Urban traffic	91%
3 Ermesinde	Urban background	89%
4 Espinho	Urban traffic	85%
5 Leça do Balio	Suburban background	89%
6 Matosinhos	Urban traffic	94%
7 Srª da Hora	Urban traffic	93%
8 Vermoim	Urban traffic	76%
9 Vila do Conde	Suburban traffic	85%
10 Vila Nova da Telha	Suburban background	96%

Figure 2.1. Study area and geographic location of the particulate matter monitoring stations in AMP, in 2004.

2.2.2. Air quality data

Exposure concentration is one of the key information required for the health impact assessment. In accordance with WHO guidelines on the Assessment and Use of Epidemiological Evidence for Environmental Health Risk Assessment (WHO, 2000; 2001), background pollution levels obtained from air quality time series should be considered to characterise the exposure concentrations. However, only three air quality monitoring points located in the study area are classified as background stations (Figure 2.1). The information obtained from these stations is not sufficient to characterise spatial variation of the background PM10 levels within the domain due to inhomogeneous pollution distribution pattern. Additionally, monitoring points classified as ‘‘traffic stations’’ could be considered for this purpose but these data should be used with caution. Traffic stations are directly influenced by the vehicle emissions in vicinity to monitoring points and provide important information on peak concentrations but their representativeness to characterise background pollution levels could be limited.

An innovative approach to obtain background pollution levels using filtering of air quality time series have been implemented in this work. It is assumed that influence of local emission sources and local dispersion conditions is presented in the time series as short-term fluctuations because temporal and spatial scales of air pollution are interrelated (Tchepel and Borrego, 2010; Tchepel et al., 2010). Therefore, decomposition of the air quality measurements on baseline and short-term components allows to remove local scale noise from the data and to improve spatial representativeness of the measurements. For this purpose, the Kolmogorov-Zurbenko (KZ) iterative filter has been used (Rao et al., 1997). The KZ(m,k) filter of the original time series x is computed as a moving average of m points applied k times (number of iterations) and is expressed as:

$$y_t = \frac{1}{m} \sum_{s=-(m-1)/2}^{(m-1)/2} x_{(t+s)} \tag{2.2}$$

The application of the KZ filter allows to decompose the original time series $C(t)$ on baseline (C^B) (deterministic) and short-term (C^S) components in time t (Rao et al., 1997):

$$C(t) = C^B(t) + C^S(t) \tag{2.3}$$

The output of the filtering process corresponds to the baseline component and the short-term component, which is defined as a difference between the original and the filtered data. The baseline component can be considered as the background concentration and the short-term represent the contribution of local emissions and dispersion conditions.

In the previous studies of air pollution time series performed in the frequency domain (Tchepel and Borrego, 2010; Tchepel et al., 2010), strong cross-correlation between urban traffic and background stations was established for PM10 fluctuations with the periodicities of about 12 h. These fluctuations are influenced by both, traffic flows and meteorological conditions. All variations of the concentrations with the period less than 12 h are influenced by local conditions and should be removed to obtain representative background concentrations. Therefore, the KZ filter was optimised to remove all the fluctuations with the periods less than 12 h from the original air quality measurements assuming the filter parameters m=3 and k=3 ($KZ_{3,3}$). The filtering approach has been applied to hourly data measured at different type of stations including urban traffic, urban background and suburban background influence. An example of the data obtained after the filtering is presented in Figure 2.2. The filter residuals defined as a difference between the original measurements and the data baseline is presented in Figure 2.3 and represent local short-term noise.

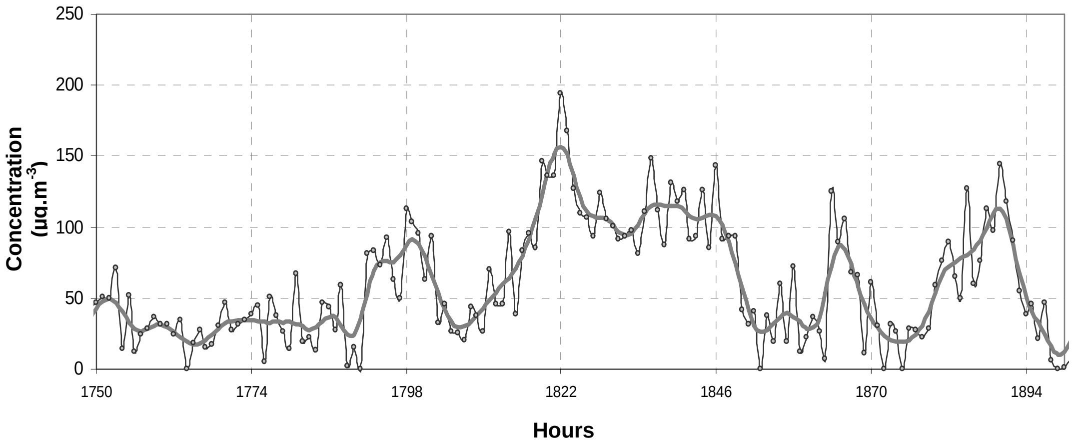


Figure 2.2. An example of PM10 concentrations before (narrow line) and after the filtering (gross line) for randomly selected hours measured in 2004 (1 year = 8784 hours) at Boavista urban traffic station.

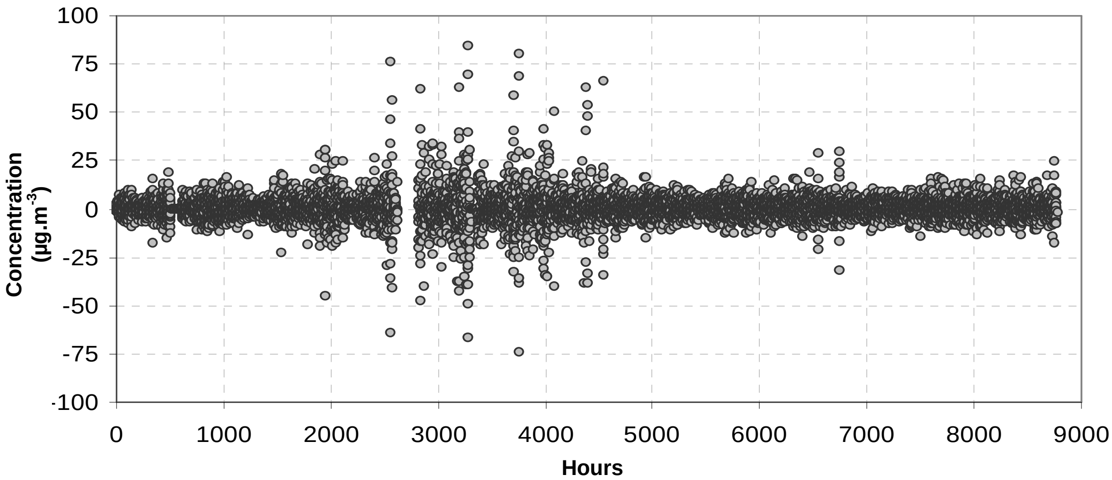


Figure 2.3. Difference between the original measurements and the filtered data (filter residual) for PM10 concentrations at Boavista urban traffic station.

The values filtered from the measurements are normally distributed with mean value of zero. The basic statistical parameters for the time series before and after the filtering are presented in Table 2.1. After the removing of local noise from the air quality time series, daily average concentrations were calculated. These data were considered in the health risk analysis together with the population mobility data to describe spatial variability of air pollution and exposed population. Alternatively, the original measurements from the background stations only (no traffic stations, without filtering) and population data on number of residents (no daily mobility) have been used. The differences between the two approaches in terms of final health benefits were investigated.

Table 2.1. Statistical parameters for annual time series considering original and filtered hourly PM10 concentrations.

Air Quality Monitoring Station	Mean	Standard deviation	Maximum
Antas			
Original	37.5	29.1	226.0
After filter	37.5	26.2	192.4
Boavista			
Original	47.5	46.0	641.0
After filter	47.5	38.6	378.2
Ermesinde			
Original	34.1	31.4	217.0
After filter	34.1	28.4	181.4
Espinho			
Original	45.1	43.8	373.0
After filter	45.1	38.4	308.7
Leça do Balio			
Original	34.2	32.7	221.4
After filter	34.2	28.7	194.7
Matosinhos			
Original	41.3	32.3	249.0
After filter	41.3	27.8	216.7
Srª da Hora			
Original	36.8	31.0	246.0
After filter	36.8	26.9	215.1
Vila do Conde			
Original	47.3	44.2	502.0
After filter	47.3	36.5	322.6
Vila Nova da Telha			
Original	35.0	28.1	240.0
After filter	35.0	24.5	193.7

2.2.3. Population mobility

Population mobility is particularly important in the studies of environmental factors that affect population health, as the level of exposure may vary substantially with geographic location (WHO, 2004). The population mobility data may provide important information on spatial and temporal distributions of inhabitants required for the exposure quantification. In this study, the data obtained from National Statistics Institute (INE, 2003) concerning daily average Origin-Destinations trips for AMP were used. One of the relevant characteristics of the study area is centralisation of working places in Porto city and an expansion of suburban zones around Porto. In all the residents of the AMP, about 28% are travelling outside the residence place, showing Porto as the main destination. Only 5% of the population are working or studying outside of AMP.

For each municipality, the mobility data together with the number of residents were used to characterise temporal and spatial variations of the exposed population (Table 2.2).

The income and outcome flows consider daily trips of the inhabitants to working or study place thus providing population distribution pattern during the daytime working hours. No distinction between working days and weekend was considered due to absence of the information. The statistical data on resident population are allocated for night-time hours. Therefore, the time of exposure and the population flows are considered to estimate daily average population exposed to inhalable particles. These population data, obtained for each municipality, are used in quantification of the number of attributable cases together with the air pollution data from a closer monitoring point.

Table 2.2. Population data considered in the health impact assessment, expressed as number of inhabitants.

Municipality	Resident population (R)	Income (I)	Outcome (O)	Daytime population (D=R+I-O)	Average population (1/2[R+D])
Espinho	31,703	2,459	3,168	30,994	31,349
Gondomar	169,239	6,015	41,073	134,180	151,710
Maia	130,254	23,964	28,403	125,816	128,035
Matosinhos	168,451	24,275	32,682	160,045	164,248
Porto	238,954	114,577	17,721	335,811	287,382
Póvoa de Varzim	65,452	3,818	6,064	63,206	64,329
Valongo	91,274	5,691	20,032	76,933	84,104
Vila do Conde	75,981	6,862	9,515	73,328	74,654
Vila Nova de Gaia	300,868	13,619	42,624	271,864	286,366

2.2.4. Health indicators, concentration-response functions (CR) and air pollution reduction scenario

Health effects of air pollution exposure are mainly related with cardiovascular and respiratory diseases (Pope et al., 1999; Dockery, 2001; Analitis et al., 2006). Therefore, the health indicators considered in this study include cardiovascular and respiratory mortality expressed as daily mortality rates in number of deaths.100 000 inhabitants^{-1} (Table 2.3).

Table 2.3. Mortality rate (number of deaths.100 000 inhabitants^{-1}) and annual mortality (number of deaths) in AMP.

Health indicator	Mortality rate (number of deaths.100 000 inhabitants^{-1})	Annual mortality (number of deaths)
Cardiovascular mortality	268.35	3166.08
Respiratory mortality	77.85	938.50

The risk of developing a disease due to exposure to agents with different levels of intensity and duration can be assessed using a statistical model for an exposure-effect relationship (Corvalan et al., 1999). Due to the absence of the information on exposure-effect relationship derived specifically for the study area, the values from epidemiological studies recommended by European study (APHEIS, 2005) were adapted as presented in Table 2.4. However, an overestimate of the Relative Risk (RR) could be expected as identified by Samoli et al. (2008). To provide a better understanding of the short-term effects of atmospheric particles on human health, two types of concentration-response functions are distinguished: (i) Effects associated with exposure to very short term (1–2 days), and (ii) the health effects due to cumulative exposure of up to 40 days (Zanobetti et al., 2002; 2003).

The health impact assessment is implemented in this study for the air pollution reduction scenario considering the legislation limit values of daily average 50 $\mu g.m^{-3}$ recently revised by the Directive 2008/50/CE and proposed in the latest review of ‘‘Air Quality Guidelines’’ from WHO (2006) as the reduction ‘‘target’’ level.

Table 2.4. Relative Risk (RR) for cardiovascular mortality and respiratory mortality associated with short-term exposure to PM10 (APHEIS, 2005). Values presented in parenthesis correspond to the 95% confidence interval (CI). Mortality rate (number of deaths.100 000 inhabitants^{-1}) and annual mortality (number of deaths) in AMP.

Health indicator	**Relative risk For 10 $\mu g.m^{-3}$ increase**	
	Very short-term (1 – 2 days)	**Cumulative short-term (40 days)**
All ages, cardiovascular mortality	1.009 (1.005 – 1.013)	1.01969 (1.0139 – 1.0255)
All ages, respiratory mortality	1.013 (1.005 – 1.021)	1.04206 (1.0109 – 1.0742)

2.3. Results and Discussion

The results obtained for short-term exposure, expressed as a number of attributable cases, are presented and discussed in this topic. Table 2.5 presents the number of annually avoided deaths due to the reduction of short-term PM10 exposure. The short-term assessment is developed for 1–2 and 40 days exposure considering cardiovascular mortality and respiratory mortality.

The results from two alternative approaches (without and with spatial variation) are compared. In the first case, average pollution concentration was calculated from the background stations and the exposed population is quantified as a total for the study area.

In the second approach, spatial variation of air pollution levels was characterised using filtered air quality time series from the 10 stations distributed within the domain and these data are used in combination with the population Origin-Destination mobility considering closest monitoring point for each municipality as described previously.

Table 2.5. Potential benefits in terms of number of ''preventable'' early deaths associated with reduction of daily mean values of PM10 to the limit value of 50 $\mu g.m^{-3}$, in AMP. Values presented in parenthesis correspond to the 95% confidence interval.

Air Pollutant Indicator	Health indicator	Potential reduction in mortality (no spatial variations in the input data)		Potential reduction in mortality considering population mobility and spatial variations of PM10 concentrations	
		Mortality rate (deaths.100 000 inhabitants^{-1})	Annual mortality (deaths)	Mortality rate (deaths.100 000 inhabitants^{-1})	Annual mortality (deaths)
Risk Assessment to Short-Term Exposure:					
PM10 very short-term (1–2 days)	Cardiovascular mortality	0.94 (0.51 – 1.36)	11.9 (6.59 – 17.3)	1.46 (0.78 – 2.03)	18.63 (9.94 – 25.83)
	Respiratory mortality	0.41 (0.16 – 0.66)	5.16 (1.97– 8.42)	0.62 (0.23 – 1.0)	7.95 (2.98 – 12.83)
PM10 cumulative short-term (40 days)	Cardiovascular mortality	2.11 (1.48 – 2.75)	26.79 (18.8 – 34.9)	3.20 (2.24 – 4.18)	40.70 (28.49 – 53.17)
	Respiratory mortality	1.41 (0.35 – 2.60)	17.97 (4.48 – 33.0)	2.12 (0.53 – 3.95)	27.03 (6.69 – 50.13)

As could be seen from Table 2.5, the results obtained from the two approaches are considerably different. The potential benefit estimated by the approach with the population mobility data is 50 – 56% higher than estimations provided by the traditional approach, revealing larger differences for very short-term exposure. This fact is related with population daily trips to the Porto city area characterised by higher pollution levels then suburbs and, therefore, resulting in higher exposure level estimated by the methodology.

As it was mentioned before, the effects of air pollution on human health depend not only on the pollutant concentration, but also on the duration of exposure of the individuals. In this context, spatial variation of the PM10 concentration and mobility of the individuals are of extreme importance. Moreover, the distinct results obtained with and without population mobility are important to analyse a sensitivity of the risk assessment methodology to the input data.

Since the methodology applied in this study for the risk assessment is based on the Apheis guidelines, a comparison of the obtained results with average European values provided by APHEIS study (2005) have been performed (Figure 2.4).

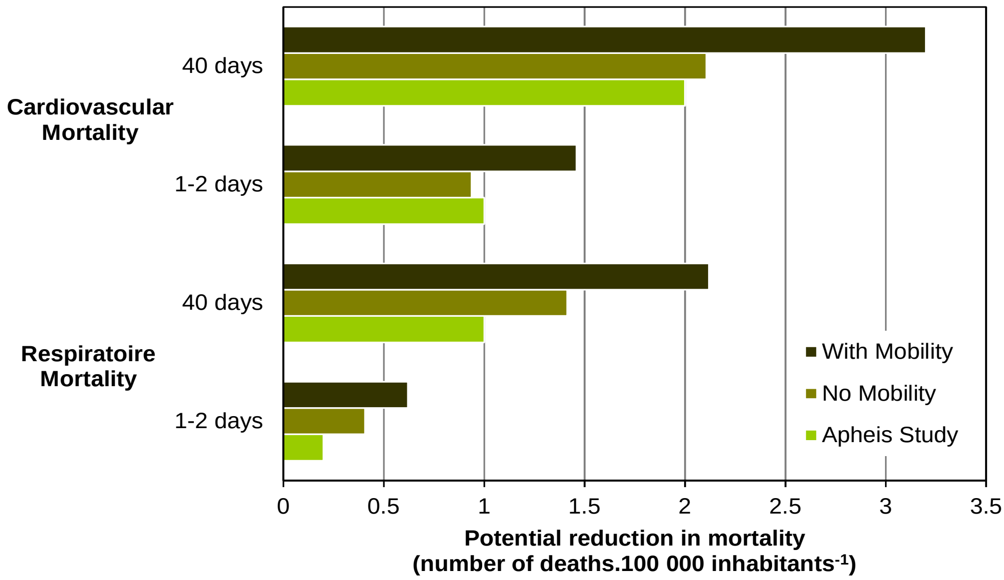


Figure 2.4. Comparison of AMP results with average European values from APHEIS study in terms of potential reductions in the number of ''premature'' deaths (number of deaths.100 000 inhabitants^{-1}).

The health benefits obtained in the current study for AMP are higher than the average European values for both indicators. The largest difference is found for potential reduction of respiratory mortality attributed to the very short-term (1–2 days) exposure achieving three times higher benefits in AMP than the average value reported for Apheis cities.

2.4. Conclusions

In this study, a quantitative assessment of potential benefits to human health related with the reduction of short-term PM10 exposure in the Porto Metropolitan Area (AMP) has been performed. High population mobility observed within the study area and the inhomogeneity of spatial pollution pattern are identified as important factors for the short-term health risk analysis. Therefore, an improved methodology to process population statistics taking into account daily average population mobility and filtering of air quality time series to improve representativeness of measurements are implemented. The methodology improves the characterisation of spatial and temporal variability in the population distribution and air pollution pattern and, consequently, the population exposure assessment. The health benefits obtained for AMP considering population mobility in the input data are 50 – 56% higher than those provided by the traditional approach and correspond to the potential annual reduction of 3.2 (95% CI 2.24 – 4.18) deaths.100 000 inhabitants^{-1} due to cardiovascular diseases and 2.12 (95% CI 0.53 – 3.95) deaths.100 000 inhabitants^{-1} due to respiratory diseases, considering cumulative short-term (40 days) exposure to PM10.

The number of annually avoided premature deaths estimated for the study area is three times higher for some health indicators than the average values reported for the European cities. However, the results are strongly influenced by the input data on population mobility and air pollution spatial variation considered in the analysis thus showing the sensitivity of the short-term risk assessment methodology to these parameters.

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CHAPTER THREE

3. PARTICULATE MATTER AND HEALTH RISK UNDER CHANGING CLIMATE: ASSESSMENT FOR PORTUGAL

Published

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Abstract
In this work the potential impacts of climate-induced changes in air pollution levels and its impacts on population health was investigated. The IPCC scenario (SRES A2) was used to analyse the effects of climate on future PM10 concentrations over Portugal and their impact on short-term population exposure and mortality. The air quality modelling system has been applied with high spatial resolution looking on climate changes at regional scale. To quantify health impacts related with air pollution changes the WHO methodology for health impact assessment was implemented. The results point to 8% increase of premature mortality attributed to future PM10 levels in Portugal. The pollution episodes with daily average PM10 concentration above the current legislated value (50 µg.m^{-3}) would be responsible for 81% of attributable cases. The absolute number of deaths attributable to PM10 under future climate emphasizes the importance of indirect effects of climate change on human health.

Keywords: air quality modelling, particulate matter, climate change, health impact assessment, mortality, Portugal.

3.1. Introduction

Climate change affects human health by a combination of direct and indirect processes. Thus, the abrupt change of temperatures leading to heat waves or cold spells has become widespread, causing fatal illnesses, such as heat stress or hypothermia, as well as increasing death rates from heart and respiratory diseases. According to the World Health Organization (WHO), the statistics on mortality and hospital admissions show that death rates increase during extremely hot days, particularly among very old and very young people living in cities. In Portugal, during the European heat wave of 2003, a total of 2,399 excessive deaths were estimated which implied an increase of 58% over the expected deaths (Trigo et al., 2009).

The indirect effects of climate change on human health are related, among others, to the changes in air pollution levels under future climate. Thus, changes in the temperature, humidity, wind, and precipitation that may accompany future climate can deeply impact air quality because of induced changes in the transport, dispersion, and transformation of air pollutants at multiple scales (Bernard et al., 2001; NRC, 2001). According to Sheffield et al. (2011), climate change could cause an increase in regional summer ozone-related asthma emergency department visits for children aged 0–17 years of 7.3% across the New York metropolitan region by the 2020s. When population growth is included, the projections of morbidity related to ozone were even larger. The authors also highlighted that the use of regional climate and atmospheric chemistry models makes possible the projection of local climate change health effects for specific age groups and specific disease outcomes.

The potential impact of climate change on particulate matter (PM) is of major concern because their concentrations are most likely to increase under a changing climate (Ayres et al., 2008; Kinney, 2008; Jacob and Winner, 2009) and because future changes in particulate matter concentrations are likely the most important component of changes in mortalities attributable to air pollution in future scenarios (West et al., 2007). Over the last few decades, human exposure to particulate air pollution has been associated with human mortality and morbidity, as well as a broad range of negative health outcomes at levels usually experienced by populations due to short- and long-term exposure to particulate matter (Künzli et al., 2000; Anderson et al., 2004; 2005; Pope and Dockery, 2006; Samoli et al., 2008; Katsouyanni et al., 2009). The European directive (2008/50/CE) revised the limit values for PM10 (particulate matter with an aerodynamic diameter less than or equivalent to 10 µm) previously defined by the Framework Directive (1999/30/EC) and set up new quantitative standards for PM2.5 (particulate matter with an aerodynamic diameter less than or equivalent to 2.5 μm). Nevertheless, PM threshold levels to which exposure does not lead to adverse effects on human health have not yet been identified and given that there is a substantial inter-individual variability in exposure and in the response, it is unlikely that any standard or guideline value will lead to a complete protection for every individual against all possible adverse health effects of particulate matter (WHO, 2006).

For Portugal, studies show frequent exceedances of EU directive targets for air quality (EEA, 2009). WHO has recently identified that Portugal is one of the 80 countries that exceed the reference values for particulate matter (WHO, 2011). In addition, particulate emissions decreased in most European countries between 1990 and 2008 except for Portugal, Bulgaria, Romania, Malta, Finland, Denmark, Latvia, and Spain, where increases were recorded (EEA, 2010). However, studies focusing on the health impacts of air quality in Portugal are very few. Several studies concerning the impact of meteorological factors on human health and the first attempt to relate air pollution levels and morbidity for Portugal have been published (Alves and Ferraz, 2005; Nogueira et al., 2005; Casimiro et al., 2006; Trigo et al., 2009; Alves et al., 2010). The authors (Casimiro et al., 2006) highlight that under future climate the meteorological conditions will be more favourable for high ozone levels (low wind speed and high temperature) that could lead to impacts on human health. Recently, a number of studies on quantitative impact assessment of air pollution on mortality in Portuguese cities have emerged (Tchepel and Dias, 2011; Garrett and Casimiro, 2011) providing information on the association of current pollution levels with adverse health effects.

The main aim of the current study is to quantify the potential impact of short-term exposure to PM10 on population health under future climate. For this purpose, climate change scenario simulated with high temporal and spatial resolution is combined with health impact assessment (HIA). Air pollution modelling for the future scenario is performed assuming no changes in the PM10 precursor emissions in comparison with the reference situation thus allowing quantification of the climate change effect independently from the other factors that affect the pollution levels. The present study provides quantitative information on forecast of the health impact attributable to air pollution under a changing climate relevant for climate change mitigation and health policies.

3.2. Methodology

The potential impact on climate-induced human health effects caused by changes in PM10 concentrations over the continental Portugal is investigated using combined atmospheric and impact assessment modelling. The study is implemented in two main steps: (i) numerical simulation of PM10 concentrations over Portugal under the IPCC SRES A2 scenario and (ii) estimation of the number of deaths attributable to the changes in PM10 levels in the atmosphere under climate change.

To quantify the health impact related with air pollution changes, the WHO methodology (WHO, 2001) was adapted and applied to the study area using the input information schematically presented in Figure 3.1.

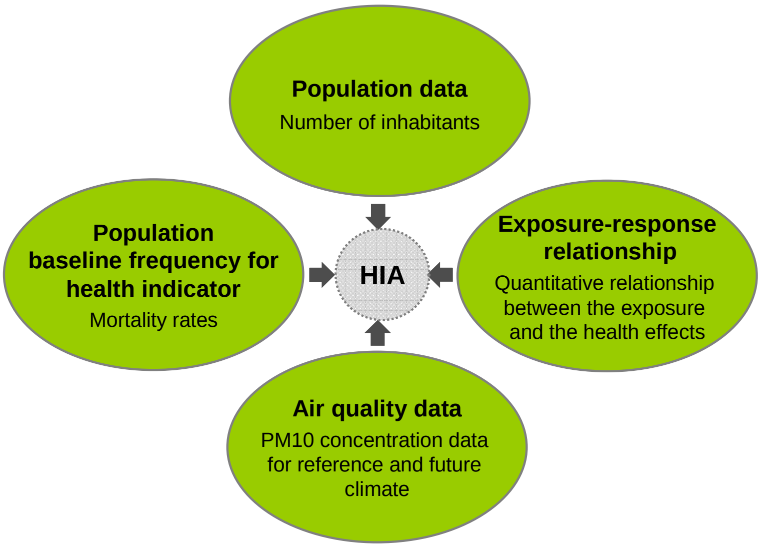

Figure 3.1. Schematic representation of the input information required by the health impact assessment performed in this study.

3.2.1. Air Quality Modelling under Climate Change

The air quality modelling was performed for a reference and a future climate scenarios first at the European scale and then over Portugal (Carvalho et al., 2010). For this purpose, global climate simulations provided by the HadAM3P model were used to drive the air quality modelling system as represented in Figure 3.2. The climate conditions for 1961–1990 are considered to characterize the reference situation, and predictions for 2071–2100 are used for the future climate in accordance with the IPCC SRES A2 scenario (Nakicenovicey et al., 2000). This scenario is considered to be the highest emission scenario and the carbon dioxide (CO_2) concentrations reaching 850 ppm by 2100. In this sense, we are assessing the worst scenario with regard to air quality changes.

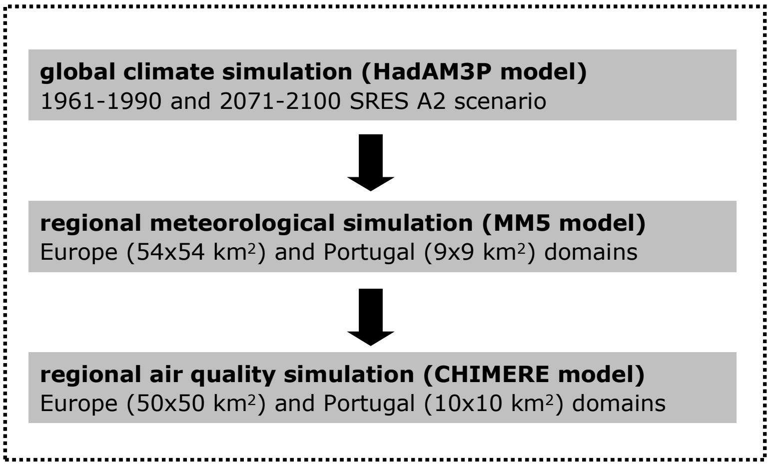

Figure 3.2. Schematic representation of the air quality numerical simulation.

The air quality modelling system is based on the chemistry transport model CHIMERE (Schmidt et al., 2001; Bessagnet et al., 2004) forced by the mesoscale meteorological model MM5 (Grell et al., 1994). The MM5/ CHIMERE modelling system has been widely applied and validated in several air quality studies over Portugal (Monteiro et al., 2005; 2007; Borrego et al., 2008) showing performance skills within the range found in several model evaluation studies using different air quality models (Vautard et al., 2007; Stern et al., 2008). TheMM5/CHIMERE modelling system has already been used in several studies that investigated the impacts of climate change on air pollutants levels over Europe (Szopa et al., 2006) and specifically over Portugal (Carvalho et al., 2010). TheMM5 mesoscale model is a nonhydrostatic, vertical sigma coordinate model designed to simulate mesoscale atmospheric circulations. The selected MM5 physical options were based on the already performed validation and sensitivity studies over Portugal (Carvalho et al., 2006) and over the Iberian Peninsula (Fernández et al., 2007). A detailed description of the selected simulation characteristics is presented in Carvalho et al. (2010). The MM5 model generates the several meteorological fields required by the CHIMERE model, such as wind, temperature, water vapour mixing ratio, cloud liquid water content, 2m temperature, surface heat and moisture fluxes, and precipitation.

CHIMERE is a tri-dimensional chemistry-transport model, based on the integration of the continuity equation for the concentrations of several chemical species in each cell of a given grid. It was developed for simulating gas-phase chemistry (Schmidt et al., 2001), aerosol formation, transport, and deposition (Bessagnet et al., 2004; Vautard et al., 2005) at regional and urban scales. CHIMERE simulates the concentration of 44 gaseous species and 6 aerosol chemical compounds. In addition to the meteorological input, the CHIMERE model needs boundary and initial conditions, anthropogenic emission data, and the land use and topography characterization. The modelling system was firstly applied at the

European scale (with 50 × 50 km^2 resolution) and then over Portugal using the same physics and a simple one-way nesting technique, with 10 × 10 km^2 horizontal resolution. The European domain covers an area from 14W to 25 E and 35N to 58N. Over Portugal, the simulation domain goes from 9.5W to 6W and 37N to 42.5N (Carvalho et al., 2006). The vertical resolution of CHIMERE model consists of eight vertical layers of various thicknesses extending from ground to 500 hPa. Lateral and top boundaries for the large-scale run were obtained from the LMDz-INCA (gas species) (Hauglustaine et al., 2005) and GOCART (aerosols) (Chin et al., 2003) global chemistry-transport models, both monthly mean values. The same boundaries conditions were used for both scenarios, since the objective is to only change the meteorological driver forcing. For the Portugal domain, boundary conditions are provided by the large-scale European simulation.

The CHIMERE model requires hourly spatially resolved emissions for the main anthropogenic gas and aerosol species. For the simulation over Europe, the anthropogenic emissions for nitrogen oxides (NOx), carbon monoxide (CO), sulphur dioxide (SO_2), nonmethane volatile organic components (NMVOC) and ammonia (NH_3) gas-phase species, and for PM2.5 and PM10 are provided by EMEP (Co-operative Programme for Monitoring and Evaluation of the Longrange Transmission of Air Pollutants in Europe) (Vestreng, 2003) with a spatial resolution of 50 km. The national inventory INERPA was used over the Portugal domain (Monteiro et al., 2007).

Reference and the IPCC SRES-A2 climate scenario over Europe and over Portugal were simulated by dynamical downscaling using the outputs of HadAM3P (Jones et al., 2005), as initial and boundary conditions to the MM5 model. The MM5 model requires initial and time-evolving boundary conditions for wind components, temperature, geopotential height, relative humidity, surface pressure, and also the specification of SSTs. Carvalho et al. (2010) discuss the global model HadAM3P and the MM5 ability to simulate the present climate. TheHadAM3P was selected to drive the MM5model because a previous work (Anagnostopoulou et al., 2008) has already concluded that the HadAM3P accurately reproduces the large-scale patterns, namely, the 500 hPa fields. The 500 hPa height reflects a broad range of meteorological influences on air quality. The authors concluded that the HadAM3P is able to capture the mean patterns of the circulation weather types. The obtained results give confidence to use the HadAM3P outputs as initial and boundary conditions for regional simulations.

To evaluate the influence of climate change on air quality, the anthropogenic emissions were kept constant (to the year 2003) in the simulations for the future climate and were not scaled in accordance with the IPCC SRES A2 scenario. This idealized regional model simulation provides insight into the contribution of possible future climate changes on the 3D distribution of particulate matter concentrations. The MM5/CHIMERE simulations were conducted from May 1st to October 30th for the reference year (1990) and for the future scenario year (2100). Both simulations had the same chemical boundary conditions. Following this methodology, it is possible to analyse the changes caused by climate change only. In Carvalho et al. (2010), a detailed analysis of the MM5/CHIMERE modelling system application under climate change has been presented and validated.

3.2.2. Population Analysis

Population size, composition, and health status were analysed for the study area as important elements required for the health impact assessment. According to National Institute of Statistics, the resident population in Portugal in 2001 was 9,869,343 inhabitants (INE, 2002). Lisbon and Porto are emphasized as the most densely populated agglomerations representing about 38% of total national population (Figure 3.3).

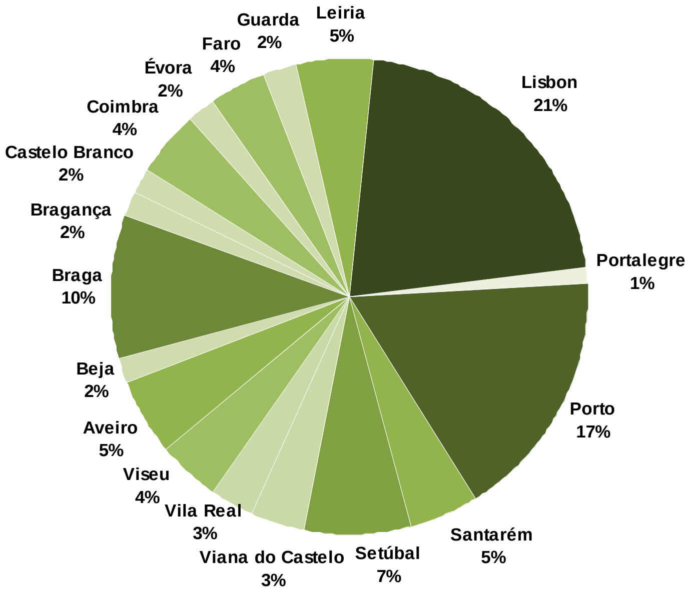

Figure 3.3. Distribution of demographic data by district in 2001.

The distribution of population by age groups is presented in Figure 3.4 stressing different proportion between active and older population for each district.

The health indicator considered in this study includes all causes mortality (except external causes) (ICD-10 codes A00-R99) expressed as daily mortality rates in the number of deaths per 100 000 inhabitants. Figure 3.5 presents the distribution of annual mortality rate by district based on DGS (DGS, 2003).

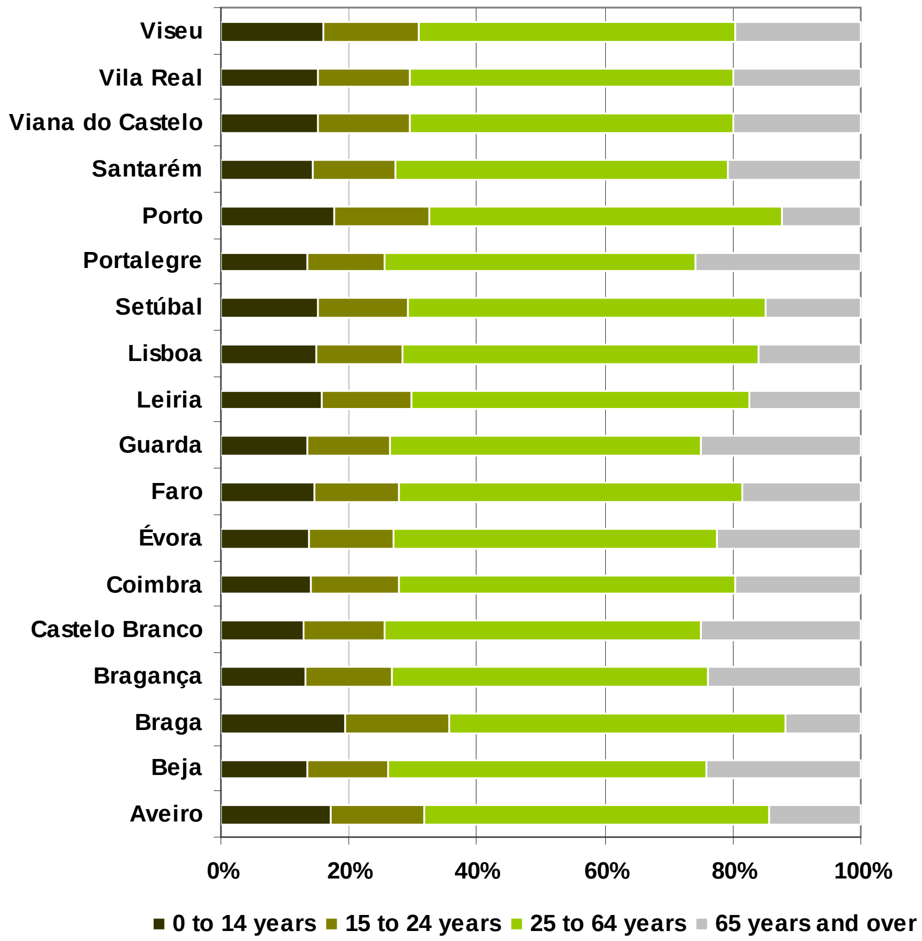

Figure 3.4. Distribution of population by age group for each Portuguese district in 2001.

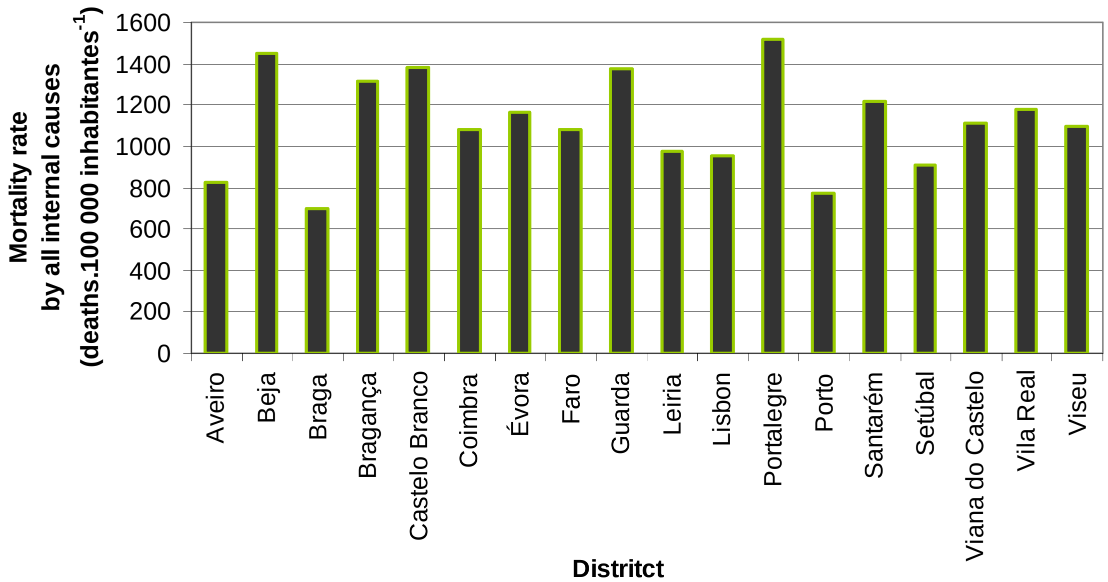

Figure 3.5. Annual mortality rate by all internal causes for each Portuguese district (deaths.100 000 inhabitants $^{-1}$) (DGS, 2003).

As could be seen, there is not a homogeneous distribution of mortality rate by the districts in Portugal. In general, the highest mortality rate by all internal causes is observed for the regions with higher proportion of older population as presented previously in Figure 3.4. Although, the Lisbon district indicates greater mortality rate than Porto with main difference in the mortality rate for age group 25 – 64 years (Figure 3.6).

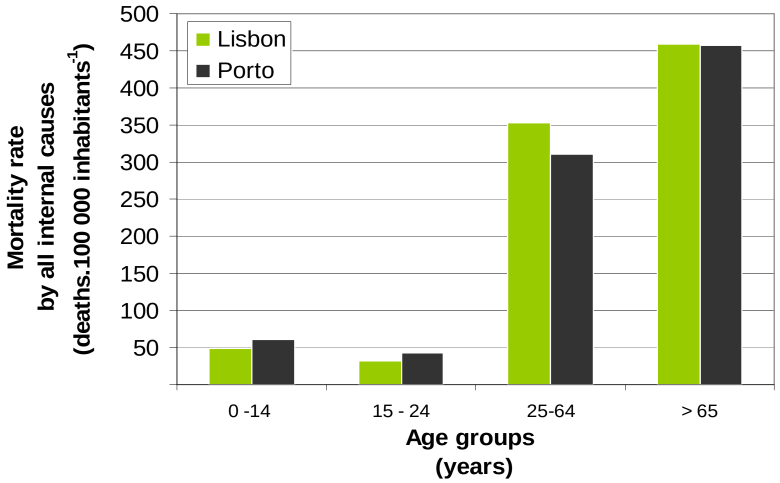

Figure 3.6. Annual mortality rate by all internal causes in Lisbon and Porto districts by age groups.

3.2.3. Health Impact Assessment

A methodology to quantify health effects is conducted in terms of number of cases attributable to air pollution that may be prevented by reducing current levels of PM10 (WHO, 2001; APHEIS, 2005). An estimate of attributable deaths (AD) is obtained from the average number of deaths ($\bar{y}$), the regression coefficient β provided by epidemiology-based exposure-response functions, and the difference between the daily average concentration ($\bar{x}$) and a reference value under a given scenario (x^*):

$$\mathrm{AD} = \bar{y} \times \beta\left(\bar{x} - x^*\right) \tag{3.1}$$

The EIS-PA model, developed by French Surveillance System on Air Pollution and Health as a support tool for automated and standardized health risk assessment (INVS, 2000), is used in this study to calculate the number of premature deaths prevented annually due to the reduction of PM to the selected “target” concentration. The results of EIS-PA model application provide estimates of the health outcomes related to short-term (1–2 days) exposure.

The exposure-response function, expressed as Relative Risk (RR) per 10.µg·m^{-3}, from epidemiological studies recommended by the European study (APHEIS, 2005) was adopted, considering the Relative Risk (RR) of 1.006 (95% CI (1.004 – 1.008)) for all-cause mortality (except external causes) to assess the effects on human health associated with the very short-term PM10 exposure (1–2 days) (WHO, 2004).

The time series of PM10 concentrations for future climate scenario together with demographic data and specific health indicators were considered in accordance with the Apheis guidelines (APHEIS, 2005) and used as input in the EIS-PA model (INVS, 2000). The health impact assessment is implemented for two air pollution scenarios: (i) a simulation for current climate (year 1990) and projected 2100 PM10 levels under the IPCC SRES A2 scenario; (ii) for the air pollution reduction scenario considering the legislation limit values of daily average 50.µg·m^{-3} recently revised by the Directive 2008/50/CE and proposed in the latest review of “Air Quality Guidelines” from WHO (WHO, 2006) as the reduction “target” level.

3.3. Results and Discussion

In this section, the estimated PM10 levels and health impact for both climate scenarios are analysed. The results obtained for short-term exposure (1–2 days), expressed as a number of attributable cases by all internal causes mortality, are presented and discussed. The increased number of attributable cases between the future and current pollution levels and the potential number of attributable cases prevented annually by reducing future PM10 concentrations to the legislation limit value (50 µg·m^{-3}) are also investigated.

3.3.1. Particulate Matter Levels under the IPCC SRES A2 Scenario

The simulated temperature increases under future climate almost reach 8.5ºC over mid and southern Europe during the warm period of May - October (Carvalho et al., 2010). These projections are in accordance to Rowell (2005) who predicted that in winter the largest warming occurs over eastern Europe, up to 7ºC, and in summer temperatures rise by 6 – 9ºC south of about 50ºN.

In Figure 3.7, an example of the projected climatic changes over Portugal is presented for July showing the largest temperature increases over the north western part of

Portugal reaching almost 10ºC. Relative humidity (RH) will decrease significantly all over Portugal. The changes in the meteorological fields (temperature, RH, wind, boundary layer) will influence the pollutants dispersion and transformation in the atmosphere.

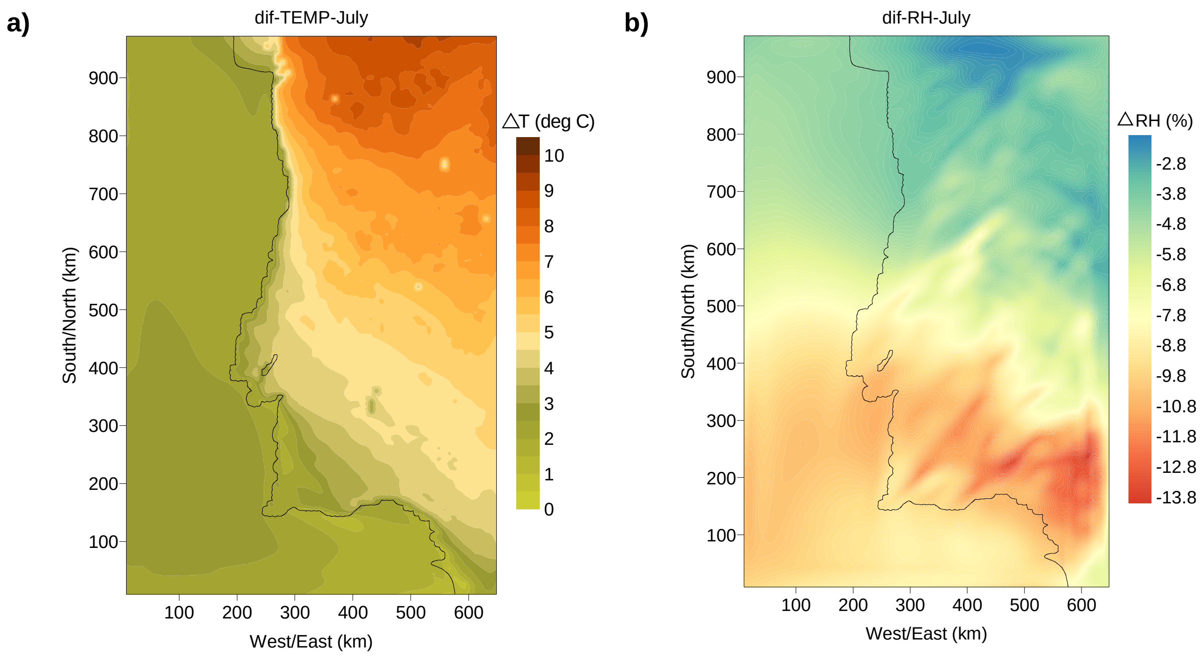

Figure 3.7. a) Temperature (ºC) and b) Relative humidity (%) differences between future and reference climates simulated with the MM5 model across Portugal for July.

Wind speed, mixing height, and relative humidity are the meteorological variables believed to mostly influence PM concentrations. Stagnant conditions are thought to correlate with high PM concentrations, as they allow particulates to accumulate near the earth's surface. Although high wind speeds can increase ventilation, they are normally correlated with high PM concentrations because they allow the resuspension of particles from the ground, as well as long-range transport of particulates between regions. High PM concentrations are normally associated with dry conditions due to increased potential to resuspension of dust, soil, and other particles. Figure 3.8 presents the average PM10 levels over Portugal over the simulation period for both climates based on hourly data provided by the air quality model.

For the overall simulation period, the maximum averaged PM10 levels increase from 60 µg.m^{-3} to 72 µg.m^{-3}. In addition, over Porto and Lisbon regions, the area affected by higher concentrations also increases in future climate (Figure 3.8).

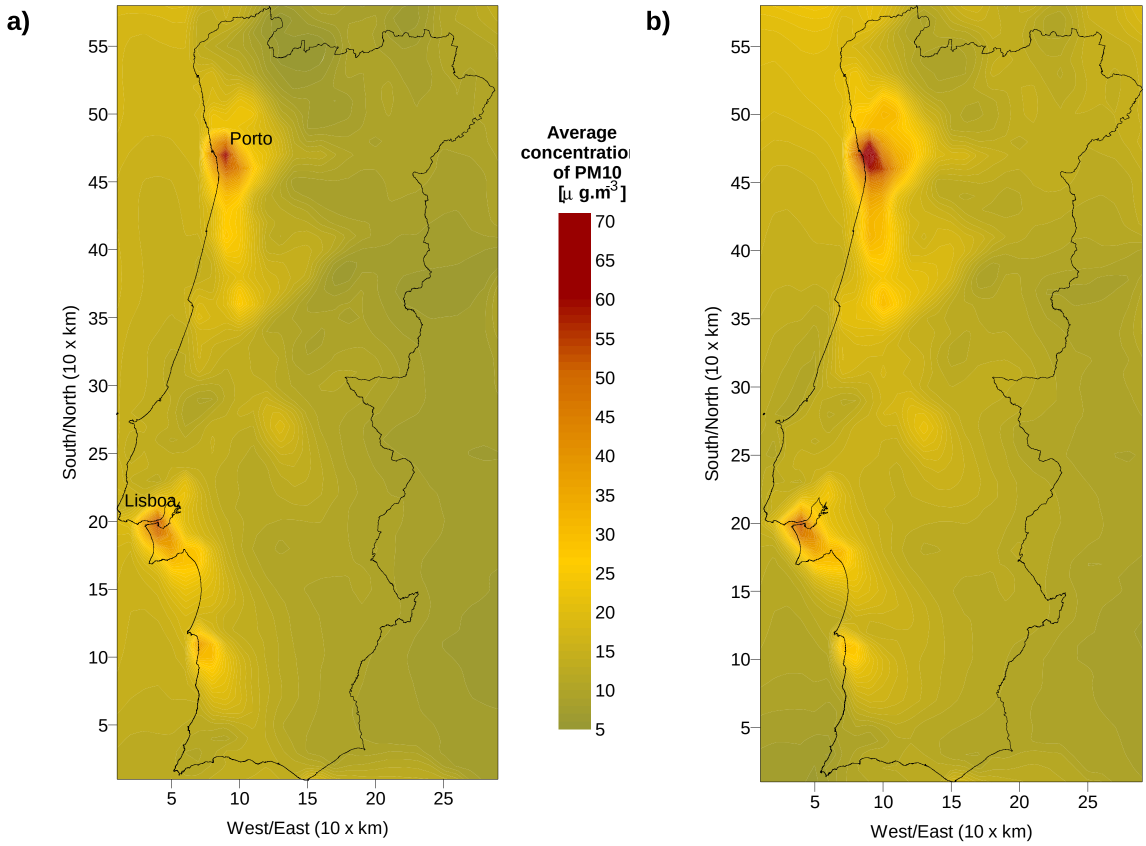

Figure 3.8. Average concentration of PM10 (µg.m^{-3}) for the simulated period (from May to October) for: a) current; b) future climate scenario.

Additionally to the changes in the average pollution levels, the frequency distribution of the PM10 concentrations is also very important for the human health studies. In Figure 3.9, an example for the most affected regions of Porto and Lisbon is presented providing information on the frequency of pollution episodes under the two climate scenarios.

The frequency distribution of the PM10 concentrations for both climatic scenarios emphasizes that Lisbon and Porto districts present an elevated number of days with high PM10 levels in comparison with the legislation limit value for the daily average PM10 concentration of 50 µg.m^{-3} that cannot be exceeded more than 35 times per year. Moreover, the climate-driven effect on PM10 levels will be more noticeable in Porto district leading to significant increase in the number of days with high daily average concentration.

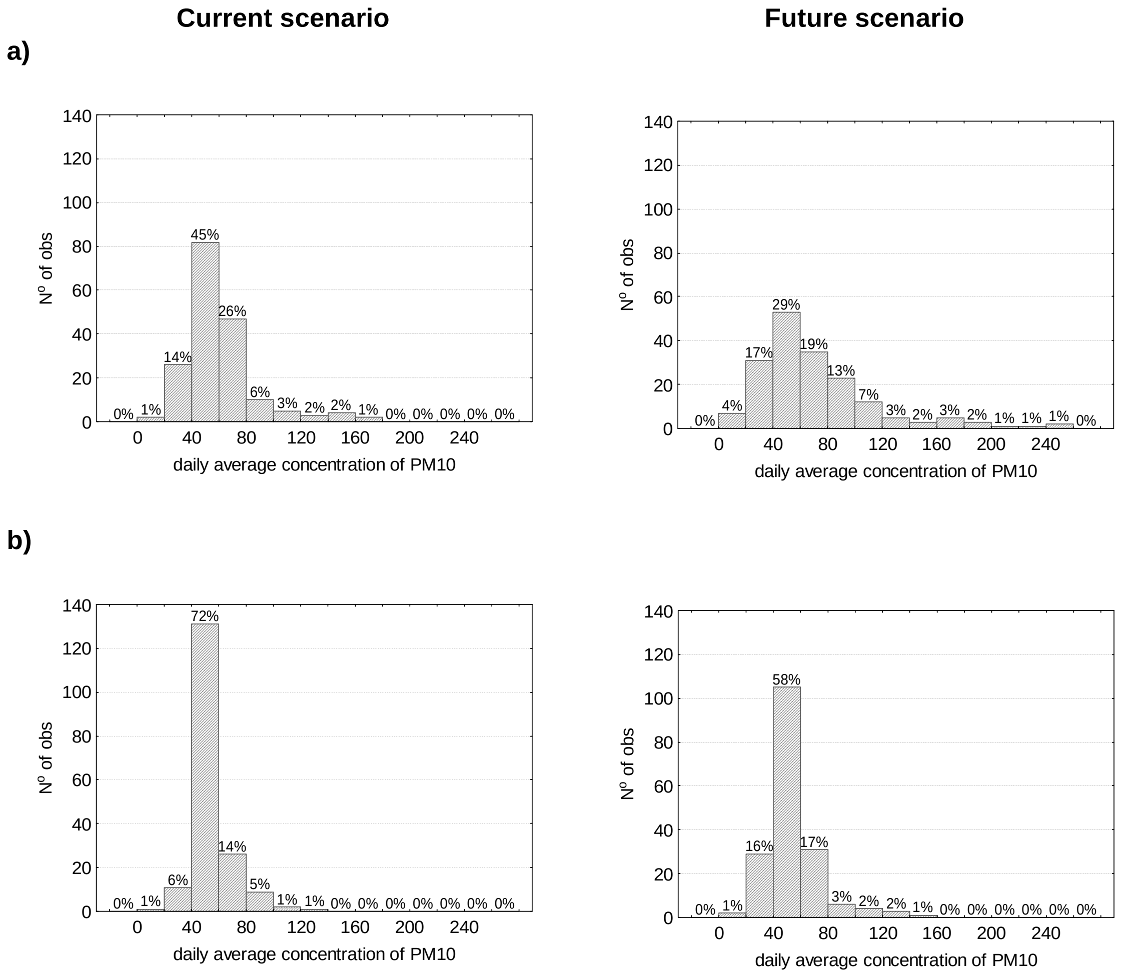

Figure 3.9. Frequency distribution of the PM10 concentrations for both climatic scenarios over the regions of: a) Porto; b) Lisbon.

3.3.2. Prognosis of Health Impact: Future versus Current Pollution Levels

The health impact assessment based on the estimated changes in PM10 between the future and reference climate shows some locations with no significant increment in the number of attributable cases to short-term PM10 exposure while other locations show important increase in PM10-induced premature mortality (Figure 3.10). Since the number of estimated attributable cases depends on both air quality and the number of the inhabitants exposed, air quality changes in the densely populated areas of the country have a greater effect than air quality changes in less densely populated areas, in general. Modelling results suggest that worsened PM10 levels will coincide spatially with many of the most densely populated areas of the country (Figure 3.8).

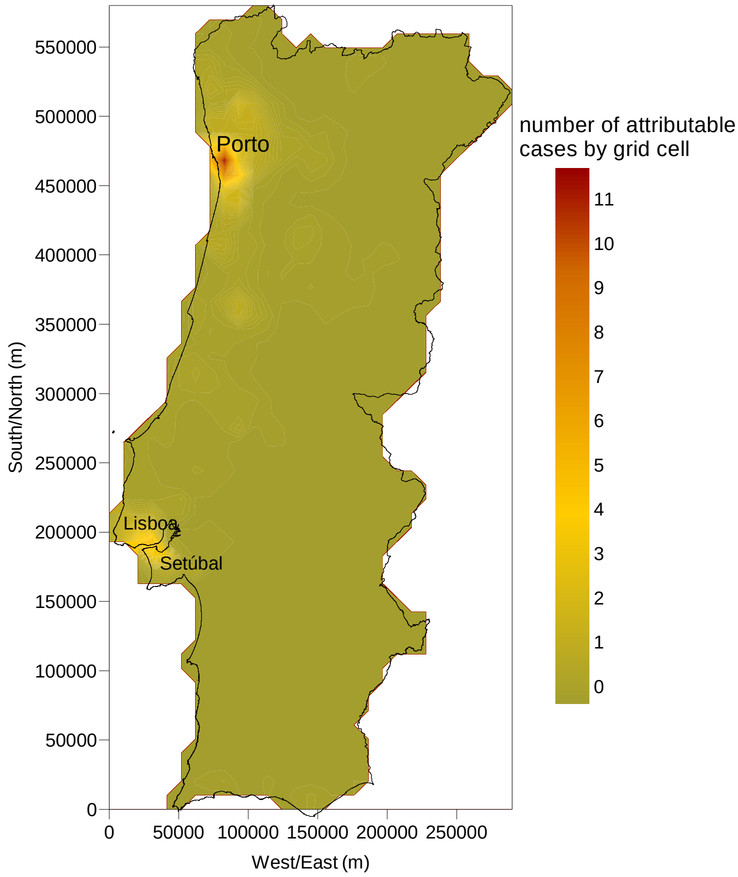

Figure 3.10. Spatial distribution of the increased number of attributable cases estimated by grid cell (10x10 km^2) related to short-term PM10 exposure for future climate.

As could be seen from Figure 3.10, the highest increase of the number of attributable cases under a future climate scenario would be expected in the Northern coastal region and Lisbon metropolitan area achieving a maximum augment of 11 cases by grid cell. The results presented in Table 3.1 highlight that the changes on the PM10 concentrations lead to a significant increase in the number of deaths in the future for most districts, especially those with the larger urban areas. Additionally, the Lisbon district is characterised by larger population size and the current mortality rate is higher, and the Porto district is the most affected (about 31% of total national deaths), reaching two times higher values than expected for the Lisbon district due to different prognosis of future pollution levels for these areas.

On the other hand, South of Portugal presents the lowest changes in the average mortality rate (Faro district: 0.9 (95% CI 0.6 – 1.2)) since the PM10 concentrations projected for 2100 will not increase significantly in comparison with the current pollution levels. At national level, about 203 (95% CI 137 – 271) more premature deaths per year are projected for 2100 in comparison to the current scenario due to indirect effect of climate change.

Table 3.1. Increase of mortality attributable to PM10 pollution levels under the climate scenario in comparison with the reference situation. Values presented in parenthesis correspond to the 95% confidence interval (CI).

District	Mortality rate average and 95% CI (deaths.100 000 inhabitants^{-1})	Annual mortality average and 95% CI (deaths)
Aveiro	2.6 (1.7 – 3.5)	13 (9 – 18)
Beja	1.7 (1.1 – 2.2)	3 (2 – 3)
Braga	1.9 (1.3 – 2.6)	19 (12 – 25)
Bragança	2.0 (1.3 – 2.6)	3 (2 – 4)
Castelo Branco	1.7 (1.1 – 2.2)	3 (2 – 4)
Coimbra	2.5 (1.7 – 3.4)	11 (7 – 15)
Évora	1.4 (0.9 – 1.9)	3 (2 – 3)
Faro	0.9 (0.6 – 1.2)	3 (2 – 4)
Guarda	1.8 (1.2 – 2.5)	4 (3 – 5)
Leiria	1.6 (1.1 – 2.2)	8 (6 – 11)
Lisbon	1.3 (0.8 – 1.7)	26 (17 – 35)
Portalegre	1.8 (1.2 – 2.4)	2 (2 – 3)
Porto	3.7 (2.5 – 5.0)	62 (41 – 83)
Santarém	1.8 (1.2 – 2.3)	8 (5 – 11)
Setúbal	1.9 (1.2 – 2.5)	13 (9 – 18)
Viana do Castelo	2.4 (1.6 – 3.2)	8 (5 – 11)
Vila Real	1.9 (1.3 – 2.6)	6 (4 – 7)
Viseu	2.0 (1.3 – 2.6)	8 (5 – 11)
National	2.1 (1.4 – 2.8)	203 (135 – 271)

3.3.3. Prognosis of Health Impact: Future Pollution versus Legislation

Additionally to the impact assessment based on prognosis of future pollution, the benefit for human health related with potential reduction of PM10 to the legislation limit value (daily average concentration of 50 μg.m^{-3}) was analysed. The number of prevented cases for all internal causes mortality attributed to the short-term (1–2 days) exposure is quantified considering that no exceedances to the limit value will occur. The results for each district are presented in Figure 3.11.

Porto district will be the greatest benefited in case of the legislated value fulfilment that is possible to achieve with implementation of additional policy measures such as emission reductions. Therefore, if no air quality exceedances will occur, about 50 premature deaths related to PM10 exposure may be avoided annually, which corresponds to four times higher values than prevented cases estimated for the Lisbon district. As expected, this fact is related with highest increase in air pollution levels predicted for Porto in future climate.

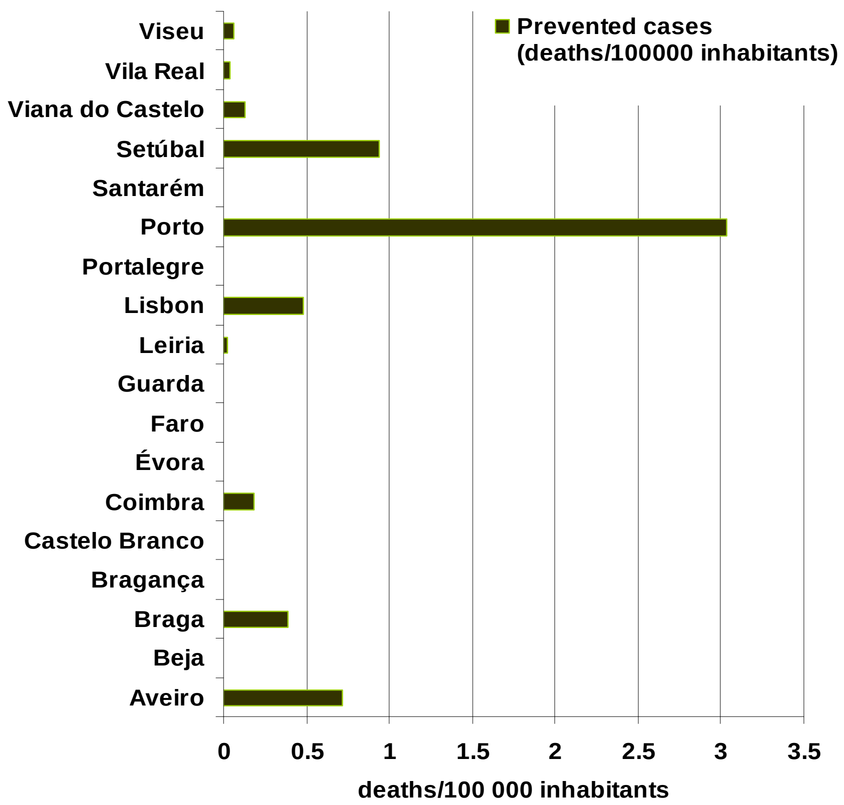

Figure 3.11. Prevented cases considering the fulfilment of the legislated value (deaths.100000 inhabitants^{-1}).

A more detailed analysis of the results obtained for the Porto area in terms of the number of attributable cases associated with different levels of exposure to PM10 is presented in Figure 3.12. Although in Porto district average PM10 concentrations above 120 µg.m^{-3} will occur in 13% of days, they are responsible for 50% of deaths attributable to air pollution (Figure 3.12). Thus emphasizing the greatest impact associated with “high pollution” days, despite their low frequency.

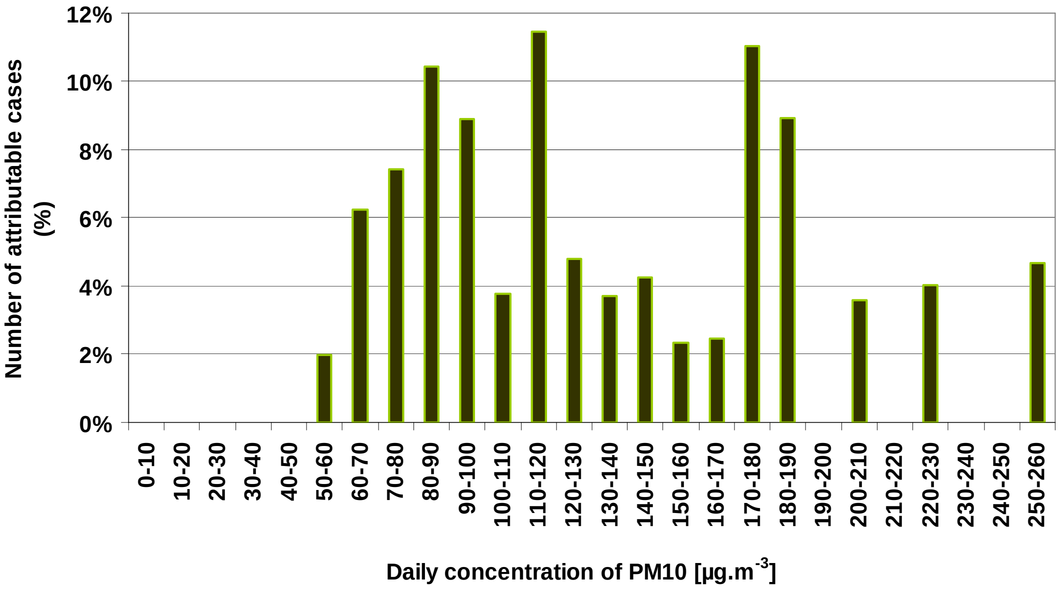

Figure 3.12. Distribution of the number of attributable cases (%) by PM10 concentration classes in Porto.

3.4. Conclusions

In this study, a quantitative assessment of the impact of climate change on human health related with short-term exposure to PM10 has been performed using combined atmospheric and impact assessment modelling. The modelling results obtained for the continental region of Portugal revealed that climate change alone will deeply impact the PM10 levels in the atmosphere. All the Portuguese districts will be negatively affected but negative effects on human health are more pronounced in major urban areas. The short-term variations in the PM10 concentration under future climate will potentially lead to an increase of 203 premature deaths per year in Portugal. The Porto district is the most affected in terms of occurrence of number of days with higher concentrations, consequently leading to the most significant increase in premature deaths that correspond to approximately 8% increase of its current mortality rate by all internal causes.

The pollution episodes with daily average PM10 concentration above the current legislated value (50 µg.m^{-3}) would be responsible for 81% of attributable cases. Although "high pollution" days have low frequency, they show the greatest impact and highlight the significant contribution of pollution peaks to acute exposure. Thus, the reduction of "high pollution" days with daily average concentration above 120 µg.m^{-3} projected to the Porto district will avoid about 50% of premature deaths attributable to air pollution.

Although the hypothetical situation of what would happen if the predicted future climate conditions will occur in 2100 and assuming that PM10 precursor emissions and population maintain constant, the information provided in this study suggests that climate-driven changes on air pollutants and human health could be substantial. Therefore, additional efforts should be made to improve on this type of modelling approach in order to support local and wider-scale climate change mitigation and adaptation policies.

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CHAPTER FOUR

4. EMISSION MODELLING OF HAZARDOUS AIR POLLUTANTS FROM ROAD TRANSPORT AT URBAN SCALE

Published

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Abstract

This study is focused on the development of a modelling approach to quantify emissions of traffic-related hazardous air pollutants in urban areas considering complex road network and detailed data on transport activity.

In this work a new version of the Transport Emission Model for line sources has been developed for hazardous pollutants (TREM-HAP). Emission factors for benzene, 1,3-butadiene, formaldehyde, acetaldehyde, acrolein, naphthalene and also particulate matter (PM2.5) were implemented and the model was extended to integrate a probabilistic approach for the uncertainty quantification using Monte-Carlo technique. The methodology has been applied to estimate road traffic emissions in Porto Urban Area, Portugal. Hourly traffic counts provided by an automatic counting system were used to characterise the spatial and temporal variability of the number of vehicles, vehicle categories and average speed at different road segments. The data for two summer and two winter months were processed to obtain probability density functions of the input parameters required for the uncertainty analysis. For quantification of cold start excess emissions, Origin-Destination matrix for daily trips was used as additional input information. Daily emissions of hazardous air pollutants from road traffic were analysed for the study area. The uncertainty of the emission estimates related to the transport activity factors range from as small as -2 to +1.7% for acrolein and acetaldehyde on highways, to as large as -33 to +70% for 1,3-butadiene considering urban street driving. An important contribution of cold start emissions to the total daily values was estimated thus achieving 45% in case of benzene. The uncertainty in transport activity data on resulting urban emission inventory highlights the most important parameter and reveals different sensitivity of the emission quantification to the input data. The methodology presented in this work allows the development of emission inventories for hazardous air pollutants with high spatial and temporal resolution in complex urban areas required for air quality modelling and exposure studies and could be used as a decision support tool.

Keywords: road traffic emissions, hazardous air pollutants, air toxics, emission modelling, emission uncertainty.

4.1. Introduction

During the last decades, road traffic has become one of the most important sources of air pollution.

Among the extended number of chemicals emitted by the vehicles, hazardous air pollutants (HAPs) require special attention due to their link with cancer and other serious adverse effects on human health. A list of 188 HAPs, referred also as air toxics, was defined in Clean Air Act by the US Environmental Protection Agency (USEPA, 2004) that contains pollutants associated with anthropogenic sources. Also, air toxics emitted by mobile sources, known as MSAT (mobile source air toxics) are identified, including: benzene, 1,3-butadiene, formaldehyde, acetaldehyde, acrolein, naphthalene and diesel particulate matter (PM) (USEPA, 2007). Emissions of MSAT are mainly related with incomplete combustion (e.g. benzene) and by-products formed during incomplete combustion (e.g. formaldehyde, acetaldehyde, and 1,3-butadiene), but evaporative processes of fuel components are also important. Besides, numerous measures to reduce air toxic emissions, including limits on gasoline volatility, limits on diesel sulphur, improvements in vehicle technology and performance, road transport is still one of the major sources of HAPs especially in urban areas. Some studies indicate that mobile sources can contribute about 68% of total HAPs emissions (Tam and Neumann, 2004). Therefore, further studies to improve quantification of air toxic emissions induced by transport in urban areas where inhabitants are living close to the pollution sources are required to better cause-effect chain analysis.

Several methodologies to quantify road traffic emissions are currently available (e.g. Zallinger et al., 2005; Smit et al., 2007; Gkatzoflias et al., 2007). However, the modelling tools not always cover HAPs or provide emissions with low temporal and spatial resolution that is not sufficient for urban scale studies. An intercomparison of the currently available models could be found at Barlow and Boulter (2009).

Urban emission inventories with higher temporal and spatial resolution are needed for a number of applications, such as urban air pollution modelling, population exposure modelling, definition of sustainable urban development policy, etc. The most commonly used technique to quantify the emissions is based upon the principle that the average emission factor for a certain pollutant and a given type of vehicles vary according to the average speed during a trip (Boulter et al., 2007a). For urban applications, hourly emissions for each road link are usually required. For this purpose, hourly traffic flows attributed to detailed road network that should be specified. Uncertainty of these data, as well as uncertainty associated with resulting emissions, is an important issue.

Quantitative methods for dealing with uncertainty in emission estimates involve the characterization of uncertainty in emission factors and/or activity data, and propagation of uncertainty to a total emission inventory. Although numerous probabilistic techniques have been applied for this purpose, the well-known Monte Carlo approach has multiple advantages and is the most often used for this purpose (e.g. Frey and Zheng, 2002a, 2002b; Abdel-Aziz and Frey, 2003). The IPCC and EPA have developed guidelines recommending the use of Monte Carlo methods as a part of a tiered approach for emissions uncertainty estimates addressing the quantification of uncertainty in emission and activity factors (USEPA, 1997; IPCC, 2000). Monte Carlo simulation methods are used to estimate uncertainty in inventories, such as for criteria pollutants, HAPs, and greenhouse gases (e.g. Winiwarter and Rypdal, 2001).

The present work intends to develop a modelling approach for quantification of traffic-related hazardous air pollutant emissions with high spatial and temporal resolution for the studies in urban areas. For this purpose, emission factors of HAPs have been implemented into the Transport Emission Model for Line Sources (TREM). Also, this new version of the model was extended to integrate a probabilistic approach for the uncertainty quantification using Monte-Carlo technique. An application example of the developed methodology to the Porto Urban Area (Portugal) for the year 2008 is presented.

4.2. Methodology

4.2.1. TREM Emissions Model

The Transport Emission Model for Line Sources was firstly developed on the basis of COST319/MEET approach and focused on carbon monoxide, nitrogen oxides, volatile organic compounds including methane, carbon dioxide, sulphur dioxide and particulate matter with aerodynamic diameter less than or equal to 10 µm (PM10) (Tchepel, 2003; Borrego et al., 2000; 2003; 2004). The prime objective of TREM is the estimation of road traffic emissions with high temporal and spatial resolution to be used in air quality modelling. Although the average-speed approach for the emission factors implemented in the model follows the European guidelines (EMEP/EEA, 2010) the way how transport activity data are considered for the emission inventorying is conceptually different. Roads are considered as line sources and emissions induced by vehicles are estimated individually for each road segment considering detailed information on traffic flow provided by automatic counting system or from a transportation model. To process these data, TREM is directly linked to Geographical Information Systems (ArcGIS) and to the transportation model VISUM (Borrego et al., 2004).

A new version of TREM developed in this work use updated emission factors from ARTEMIS methodology (André and Joumard, 2005; Boulter et al., 2007b). Following the definition of air toxics relevant for mobile sources, this new version TREM–HAP (Transport Emission Model for Hazardous Air Pollutants) is prepared to calculate the emissions of benzene, 1,3-butadiene, formaldehyde, acetaldehyde, acrolein, naphthalene and also particulate matter with aerodynamic diameter less than or equal to 2.5 µm (PM2.5). The calculation algorithm is schematically represented in Figure 4.1.

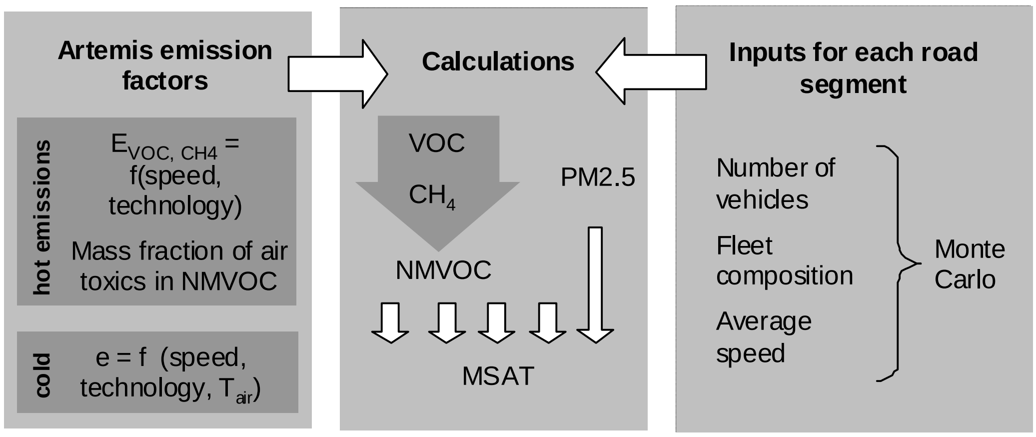

Figure 4.1. Calculation algorithm for hazardous air pollutants implemented in TREM–HAP model.

Firstly, exhaust hot emissions of total VOC, Methane (CH_4) and PM2.5 are estimated as a function of average speed for each class of vehicles. Both total emissions under thermally stabilised engine and additional cold-start emissions are considered due to the importance of cold-engine driving within urban areas. At next, methane hot emissions are subtracted from VOC and nonmethane VOC (NMVOC) emissions are separated into different compounds, including hazardous pollutants, using %-fractions as proposed by EMEP/ EEA (2010) guidelines. MSAT cold start emissions are estimated as a function of average speed and ambient temperature. In this case, passenger cars only are considered due to the methodology limitations. An example of hot exhaust emission factors calculated for benzene and formaldehyde for different type of vehicles as a function of average speed is presented in Figure 4.2 for Euro 2 technology.

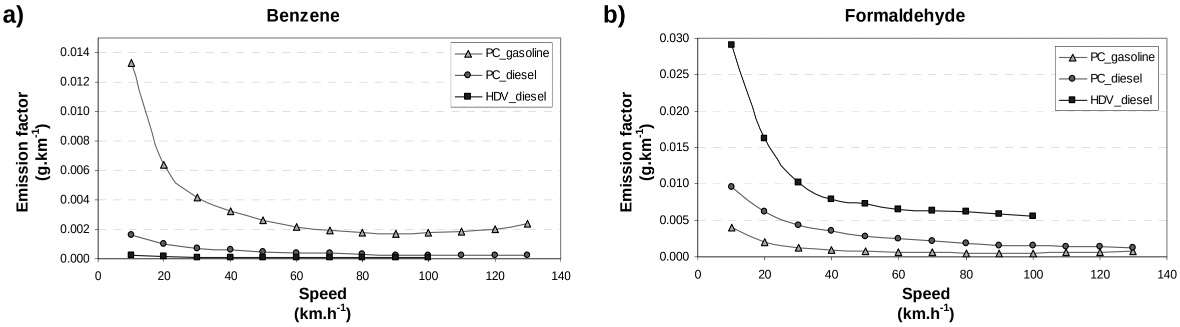

Figure 4.2. An example of emission factors for a) benzene and b) formaldehyde considered by the emission model for Euro 2 vehicles (PC_gasoline – passenger gasoline cars; PC_diesel – passenger diesel cars with engine capacity < 2 ltr; HDV_diesel – heavy duty diesel vehicles < = 7.5 t).

4.2.2. Hot Emissions

The hot emission of the pollutant p (E_p (g)) for each road segment is estimated by the model as following:

$$E_p = \sum_i \left(e_{ip}(v) \cdot N_i \right) \cdot L \tag{4.1}$$

where $e_{ip}(v)$ is the emission factor (g.km^{-1}) for pollutant p and vehicle class i defined as a function of average speed v (km.h^{-1}); N_i is the number of vehicles of class i and L is the road segment length (km).

The emission factors depend on average speed, fuel type, engine capacity and emission reduction technology. However, these data are not available for each counting point and statistical information is usually used to characterise vehicle fleet composition. In this context, uncertainty estimation of the resulting emissions became an important issue.

4.2.3. Cold-Start Emissions

Cold-start emissions are emitted by vehicles under cold engine and are estimated as an excess to the stabilised hot emission levels. The cold-start excess emission is defined as a difference between the total amount of the pollutant emitted between the start time (t = 0) and time t_{cold}, and the amount of pollutant which would be emitted by the vehicle at its normal running temperature during the same time period. Travel distance, average speed and ambient temperature are considered to quantify cold-start emissions for different vehicle technologies. At urban scale, travel distance is often less than the distance necessary to warm up the engine. Therefore, cold emissions are playing a very important role and their contribution to the total emissions could not be neglected.

In this work, the methodology developed by ARTEMIS (André and Joumard, 2005) was adapted in order to be compatible with the model conception. For this purpose, original emission factors represented as absolute emissions (g) per cold cycle were transformed to average cold emission factors (g.km^{-1}) within cold distance.

Cold emission factors are calculated as following:

$$e_{cold} = w_{20^{\circ}C,20\ km/h} \cdot f\left(T,V\right) \cdot h\left(\delta\right) \cdot g\left(t\right) \qquad (4.2)$$

where e_{cold} is the excess emission with a cold engine for a trip (g); V is the average speed during cold engine regime (km.h^{-1}); T is the ambient temperature (ºC); $h(\delta)$ is the distance correction factor = distance travelled (d) / cold distance (d_{cold}) (dimensionless); $w_{20^{\circ}C,\ 20km/h}$ is the excess emissions at reference conditions for T = 20ºC and V = 20 km.h^{-1} (g); $f(T,V)$ is the correction factor for speed (V) and temperature (T) effects; $g(t)$ is the correction factor for the parking time t.

The ARTEMIS methodology to calculate cold distance was used in order to determine the distance necessary to warm up the engine and to stabilise emissions. A schematic representation of the effect of trip length on the emissions for different classes of passenger cars is presented in Figure 4.3. As could be seen in the Figure 4.3, the emissions will stabilize within the first 5–10 km after the start that is considered as a “cold distance”.

The ARTEMIS methodology used to calculate cold-start emissions is available for passenger cars only, because of insufficient data for other categories, and for typical urban driving, which imply that only urban roads were considered (see Section 4.3.2). The input parameters considered in the determination of cold-start emission factor are presented in Table 4.1 considering different passenger car emission classes and fuel type. Calculation of the cold-start emission factor is dependent to the ambient temperature and average speed. The calculation algorithm for acetaldehyde, acrolein and formaldehyde is not sensitive to the ambient temperature. In addition, it should be noted that 1,3-butadiene emissions are totally attributed to gasoline vehicles, while PM2.5 is mainly related with diesel engines.

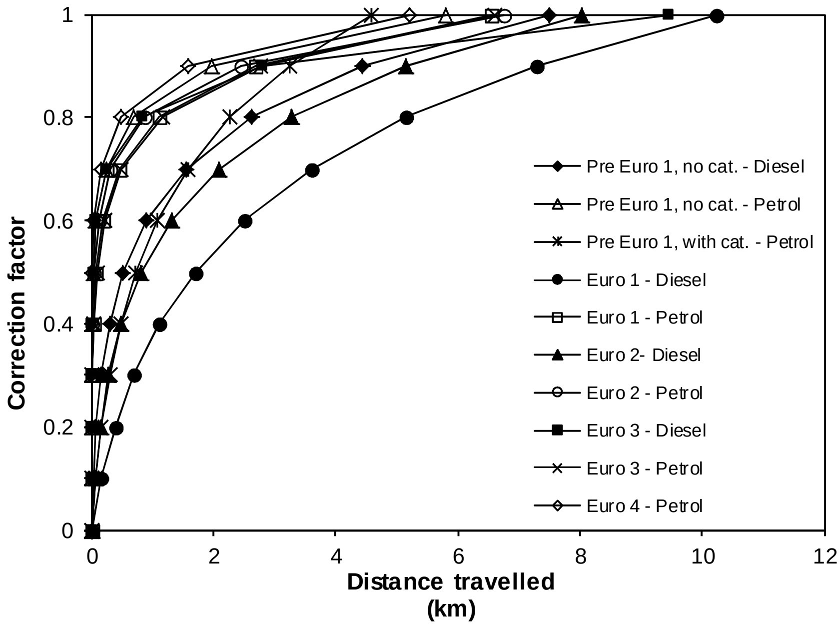

Figure 4.3. Schematic representation of the effect of trip length on the cold start excess emissions from passenger cars in winter season.

In case of naphthalene, the methodology applied is different and is not presented in Table 4.1 since the hot and cold emissions are calculated simultaneously and cannot be distinguished.

Table 4.1. Parameters considered for cold-start and hot emission factor quantification.

Pollutant	Passenger cars									
	Pre Euro 1		Euro 1		Euro 2		Euro 3		Euro 4	
	gasoline	diesel	gasoline	diesel	gasoline	diesel	gasoline	diesel	gasoline	diesel
PM2.5	–	T	–	T	–	T	–	T	–	T
Acetaldehyde	V	const.	V	const.	V	V	V	V	V	V
Acrolein	–	const.	–	const.	const.	const.	const.	const.	const.	const.
Benzene	V,T	const.	const.	const.	V,T	V,T	V,T	V	T	V
1,3-Butadiene	V	–	const.	–	V	–	V,T	–	T	–
Formaldehyde	V	const.	V	const.	V	V	V	V	V	V

Notes: T: Ambient temperature (ºC); V: Average speed ($km.h^{-1}$); const.: constant value; – : methodology not available

4.2.4. Monte Carlo Approach

The Monte Carlo (MC) approach is used to analyse uncertainty propagation, where the goal is to determine how variations in input data affect the emission estimations. For this purpose, a probability distribution is specified for each model input based upon statistical analysis of data. At next, random values are generated for each input parameter taking into account their probability distribution and assuming that the generated values represent real world events. Multiple runs of the emission model based on stochastic inputs provide multiple outputs that can be treated statistically as if they were an experimentally or empirically observed set of data, instead of obtaining a single number for model outputs as in a deterministic simulation (Frey and Bammi, 2002).

In the present work, the emissions model has been adapted to use multiple set of randomly generated values for each of the input parameters that characterise the transport activity. Thus, random samples of the number of vehicles, average speed and fleet composition are generated from the respective Probabilistic Density Functions (PDF) and one random value for each input is entered into the model to arrive at one estimate of the model output. This process is repeated over more than 600 iterations to arrive at multiple estimates of the model. These estimates are sample values of the PDF of the model output that reflects the uncertainty in the model inputs.

4.3. Application

4.3.1. Study area

The Porto Urban Area was selected in this study to quantify road traffic emissions of hazardous air pollutants. It is the second largest city in Portugal with a total area of approximately 41 km^2. The resident population of this urban area in 2008 is about 216 000 inhabitants (2% of the national population). One of the relevant characteristics of the study area is the centralisation of working places in Porto city centre and an expansion of the agglomeration around the city showing the importance of the population home/work daily trips and consequent air pollution problems in the Region (Tchepel and Borrego, 2010).

To study atmospheric emissions induced by transport, the road network was subdivided into 3 types: urban streets, interurban roads and highways with the total length of 78.3 km, 29.8 km and 22.3 km respectively (Figure 4.4a). As a total, 84 points distributed within the domain were considered to characterize traffic volume fluctuations. For this purpose, traffic data collected by automatic measurements during winter (January and February) and summer (July and August) periods of 2008 were attributed to the road links using road classification and the proximity criteria.

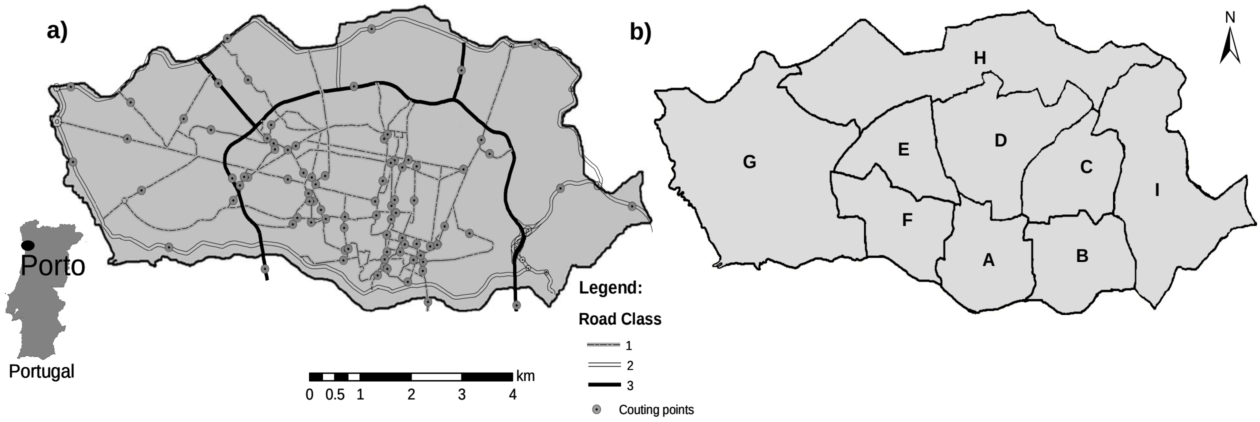

Figure 4.4. a) Administrative limits of the Porto Urban Area and road network considered in the study (type 1 – urban streets, type 2 – interurban roads, type 3 – highways); b) sectors limits considered in the O/D matrix.

Additionally, population mobility data concerning Origin/Destination trips for traffic peak hours (Oliveira et al., 2007) was considered for the study area and subdivided in 9 sectors (Figure 4.4b, Table 4.2). These statistical data provide important information for quantification of cold start emissions as described in Section 4.3.2.

Table 4.2. Origin/Destiny Matrix for each sector (number of displacements in individual transport) for the morning traffic peak period (7h30 – 9h30) (Oliveira et al., 2007).

OD Matrix	A	B	C	D	E	F	G	H	I	Ext. South	Ext. North	Total
A	269	461	430	1,070	565	445	500	523	265	447	1,819	6,794
B	315	84	357	398	200	108	98	275	168	163	504	2,670
C	569	436	304	587	344	299	379	622	265	248	587	4,640
D	879	335	676	869	609	653	758	902	198	419	1,498	7,796
E	603	136	391	526	329	532	730	291	103	106	512	4,259
F	500	159	198	431	302	215	779	281	47	170	499	3,581
G	1,344	300	353	774	859	1,255	406	1,298	135	663	2,527	9,914
H	855	445	795	1,053	639	672	652	582	325	456	1,603	8,077
I	371	396	383	416	208	204	138	265	100	81	319	2,881
Ext. South	1,686	998	810	1,542	1,093	1,427	906	735	382	8	14,400	23,987
Ext. North	7,168	2,198	3,280	3,737	2,166	4,127	4,493	4,549	1,208	11,455	11,021	55,402
Total	14,559	5,948	7,977	11,403	7,314	9,937	9,839	10,323	3,196	14,216	35,289	130,001

4.3.2. Input Data

In order to characterize the uncertainty in input parameters, a set of random inputs characterizing the fleet composition, traffic flow and vehicles speed are generated for each road. The PDF for vehicle classes is determined using the statistical information on vehicle registers and average number of kilometres travelled. For the traffic volume, data from the counting points attributed to each link were used, describing both temporal and spatial variations (Figure 4.5). Due to absence of vehicles speed measurements, this variable is estimated for each road segment considering the type of the road and taking into account the speed traffic behaviour adapted from Joumard et al. (2007): urban (30±9.4 km.h^{-1}), interurban (70±17.6 km.h^{-1}) and highways (110±8.8 km.h^{-1}). A combination of random values generated by the Monte Carlo approach is used to create 625 independent inputs for each road segment to be used by TREM–HAP for the emission estimations.

To estimate excess cold start emissions, a number of vehicles with cold engine have to be considered for each urban road segment. However, it is not possible to obtain this information directly from the automatic traffic counts that is why additional information is required. For this purpose, the ARTEMIS methodology (André and Joumard, 2005) to calculate cold distance was used in order to determine the distance necessary to warm up the engine and to achieve a constant emission level (Figure 4.3). The statistical information on Origin–Destination (O–D) mobility (Oliveira et al., 2007) was considered to determine the daily number of cold starts and the distance between the origin and destination points. Stop duration of 7 hours between the morning and evening peak hours was assumed to calculate the correction factor for cold start emissions. Based on this information, the number of vehicle × km performed with a cold engine and a proportion of cold/hot driving was calculated for each urban zone and attributed to the road network.

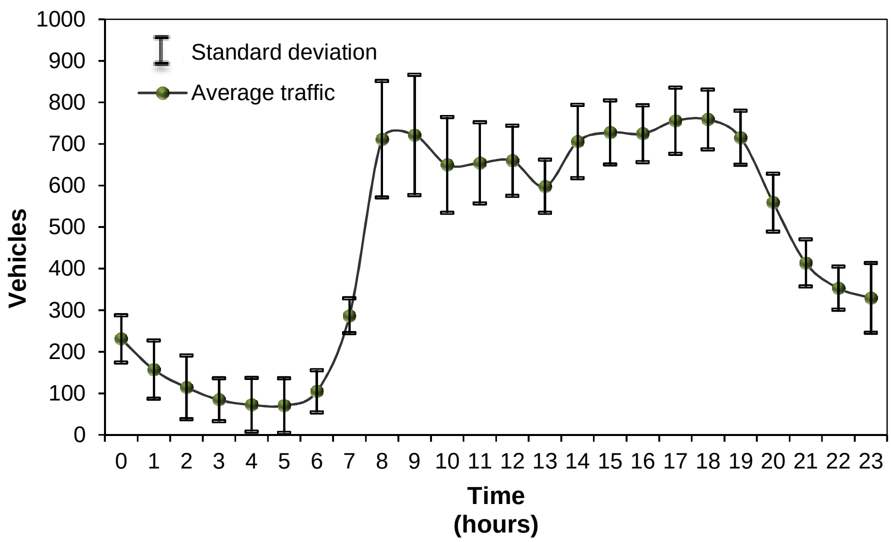

Figure 4.5. An example of temporal variation of the passenger car flows obtained from the automatic counting data at a fixed point.

4.4. Results and Discussion

The probabilistic emission inventory for the mobile source hazardous air pollutants was developed based on probabilistic activity factors. It should be stressed that uncertainty of the emission factors was not considered in the current simulations due to absence of the information. Therefore, the overall uncertainty of the emissions is related to the uncertainty in activity data only. The analysis of results examines the influence of the seasonal variations (summer and winter periods), the contribution of hot/cold start to the total daily emissions, the differences of road types and the spatial distribution of the total emissions over the study domain.

The absolute values for total daily emissions estimated for the Porto Urban Area are presented in Figure 4.6. Several statistical parameters, including average emissions, 5th and 95th percentile and extreme values were analysed for the selected hazardous pollutants. Also, seasonal difference between summer and winter are examined. It is apparent that PM2.5 and benzene have the largest absolute uncertainty in the daily emissions. For all the pollutants, except benzene, the absolute values for total daily emissions are larger in summer. Benzene has a different seasonal behaviour because of the important contribution of cold start emissions as observed in Figure 4.7.

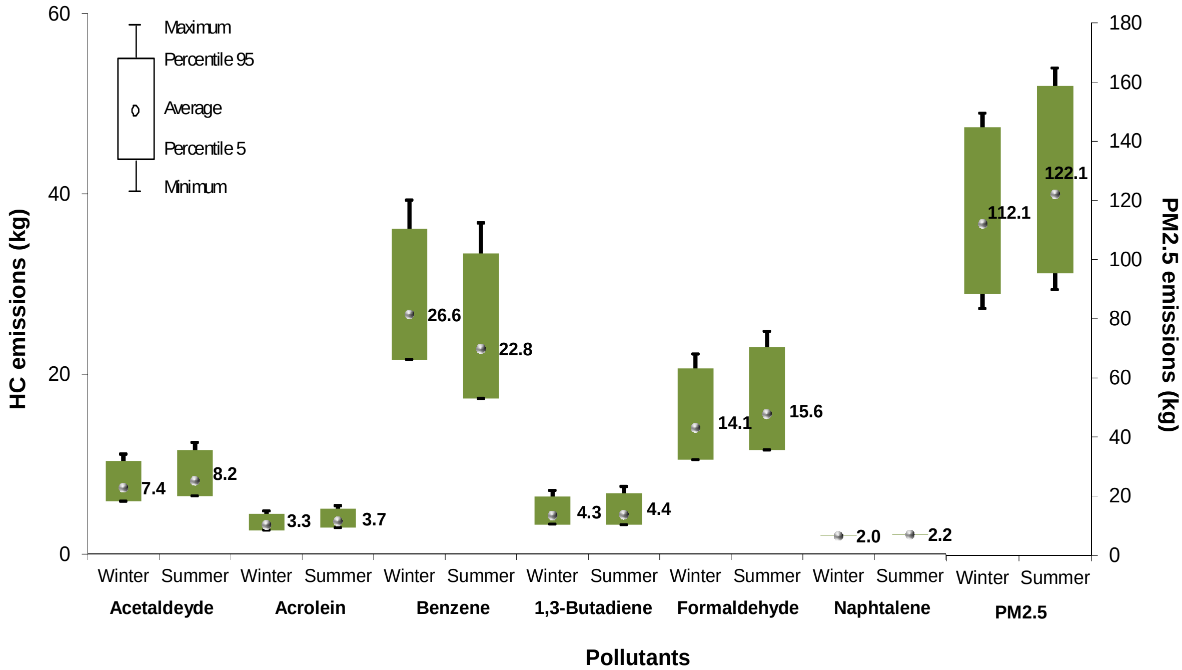

Figure 4.6. Statistical parameters for total daily emissions in the Porto Urban Area considering winter and summer periods.

The 90% probability range of the emission estimates are given in Table 4.3 considering different types of roads. For all the pollutants, urban streets are characterised by higher uncertainty in the emissions achieving the largest range for 1,3-butadiene (-33% to +70%), while estimations for highways are more robust. Benzene emissions from urban roads are less uncertain than other hydrocarbons, except naphthalene, due to the important proportion of cold start emissions with lower sensitivity to the input data. The very low uncertainties obtained for naphthalene are explained by the different methodology applied for this pollutant. The hot and cold emissions are calculated simultaneously and cannot be distinguished. Also, the methodology to calculate naphthalene emissions is not sensitive to ambient temperature and speed.

Table 4.3. Results of the uncertainties in the emission rates (hot+cold) for the different types of roads.

Pollutant	90% probability range of the emission estimates (%)*					
	Urban streets		Interurban roads		Highways	
	(-)	(+)	(-)	(+)	(-)	(+)
PM	-28.1	44.7	-11.6	30.9	-15.7	8.8
Acetaldehyde	-24.7	50.7	-16.3	28.1	-2.0	1.7
Acrolein	-26.6	53.2	-14.9	25.3	-2.0	1.7
Benzene	-22.6	43.4	-23.7	40.2	-5.3	6.1
1,3-Butadiene	-33.2	70.4	-21.3	36.5	-3.0	3.5
Formaldehyde	-36.8	65.7	-16.7	28.6	-2.1	2.0
Naphtalene	-0.7	0.6	-0.7	0.6	-0.8	0.7

* (-) =(5th percentile-Mean)/Mean) x 100;   (+) =(95th percentile-mean)/Mean) x 100

The contribution of cold emissions to the total emissions estimated in the study area at typical summer and winter days is presented in Figure 4.7.

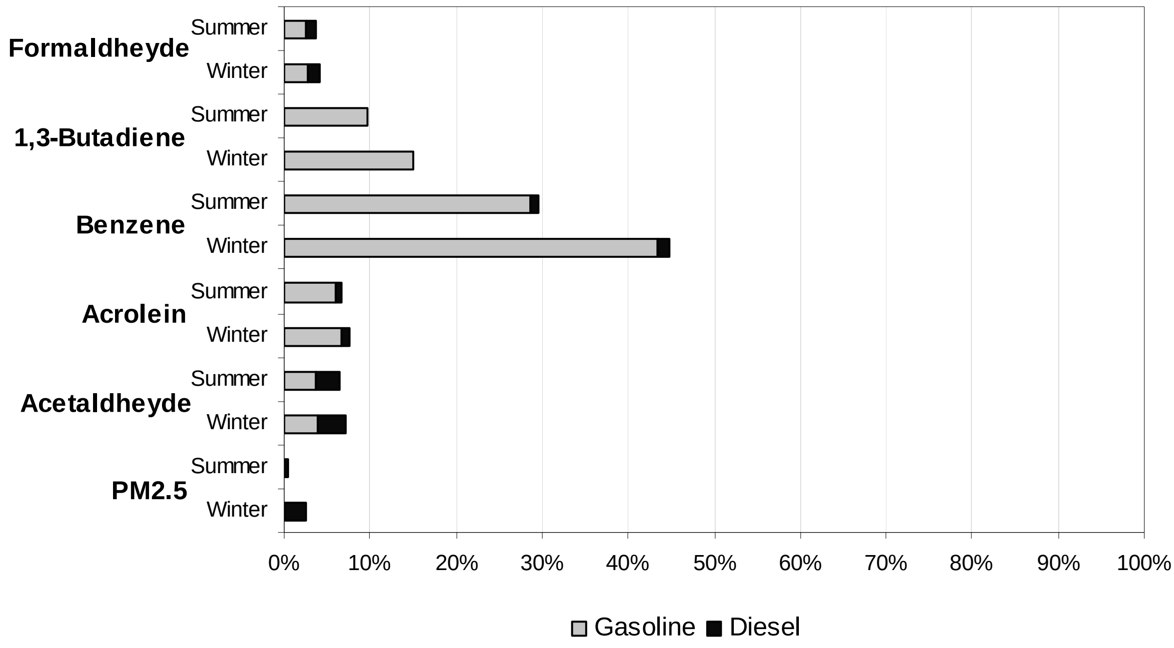

Figure 4.7. Contribution of the cold start emission (average values, percentage) to the total emissions within the modelling domain.

The results show that the contribution of cold start emissions to the total values calculated for the urban area can achieve 45% in case of benzene, while for other hazardous pollutants this contribution is below of 10% with the only exception of 1,3-butadiene. In general, excess cold start emissions from diesel vehicles are less significant compared with those from gasoline vehicles. As expected, the cold emissions are higher in winter than in summer season due to the direct influence of ambient temperature. However, in the case of acetaldehyde, acrolein and formaldehyde this difference is related to traffic fluctuations only because the calculation algorithm for these pollutants is not sensitive to the ambient temperature. It should be noted that 1,3-butadiene emissions are totally attributed to gasoline vehicles, while PM2.5 is mainly related to diesel engines.

Additionally, the spatial distribution of the daily emissions (hot + cold) was analysed for the study area. Examples for benzene and PM2.5 are presented in Figure 4.8. A different spatial pattern is observed for these two pollutants. Within the Porto Urban Area the highest emission rates of PM2.5 are estimated for highways due to intense traffic during the day.

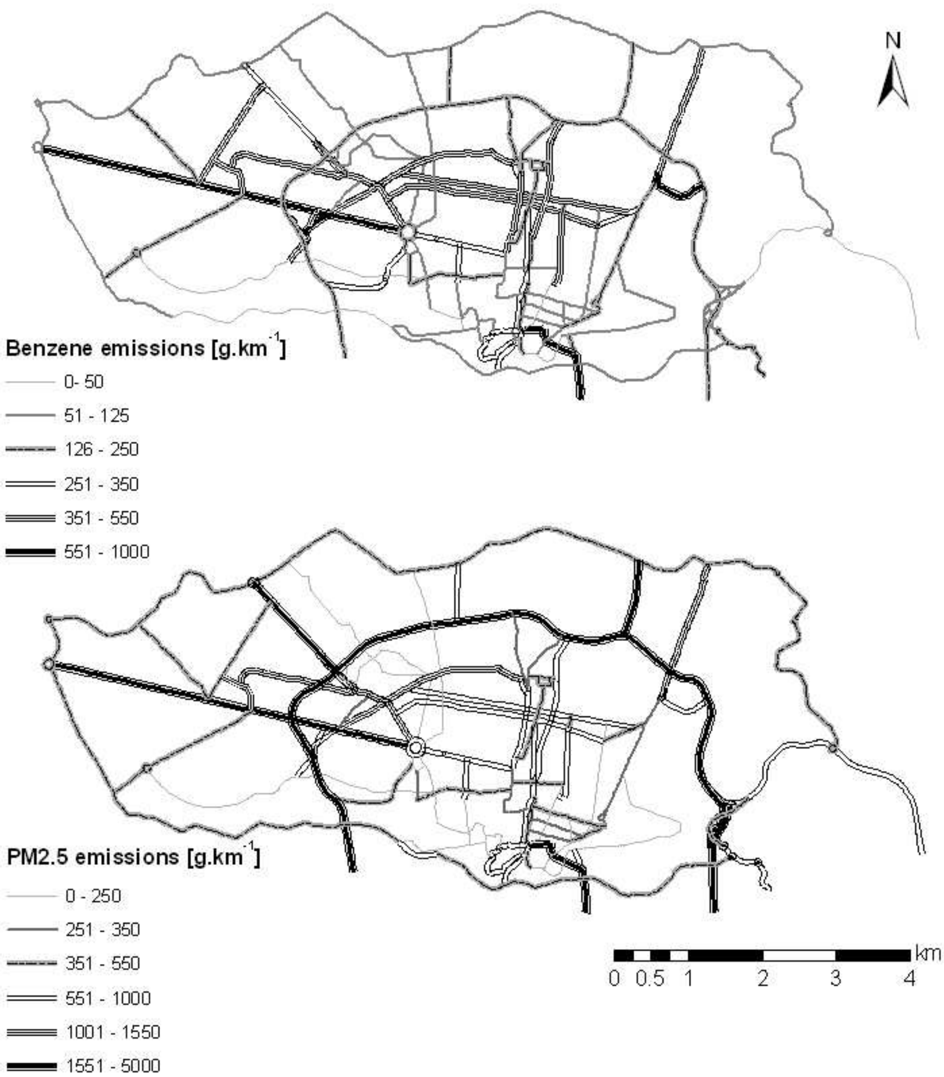

Figure 4.8. Spatial distribution of benzene and PM2.5 daily emissions (average) in the modelling domain.

Oppositely, benzene emissions are more pronounced at urban streets where the contribution of cold start emissions is very important. For both pollutants, high emissions are obtained in two urban roads which are important thoroughfares connecting the urban centre with peripheral interurban and highway roads.

4.5. Conclusions

The TREM-HAP model has been developed to estimate the emissions of hazardous air pollutants related to the traffic activity in urban areas. The current work provides a description of the methodology and an application example to characterise a probable distribution of the emissions for different types of roads considering vehicle technology mix, driving conditions and traffic volume fluctuations.

The total daily emissions of air toxics are presented for the entire study area considering their seasonal variations. Different trend is identified for benzene showing 17% higher emissions at winter time due to important contribution of cold starts while other toxic pollutants are mainly affected by changes in the traffic volume that results in higher emissions during the summer period.

Highly uncertain emission data are obtained for the urban roads with the largest range for 1,3-butadiene (–33% to +70%). Oppositely, emissions calculated for highways are generally characterised by a very small uncertainty (less than ±5%) except for PM2.5 (–16% to +9%).

The study shows that cold-start emissions can contribute up to 45% to the total daily emissions, highlighting the importance of accounting for cold start emissions in a traffic-related emissions inventory development.

Globally, the results demonstrated that the range of the uncertainty produced in the model application depends on uncertainties in the model inputs but sensitivity of the modelling approach is different for the considered air toxics.

The modelling tool developed and applied in the present work provides spatial distribution of the air toxic emissions for urban areas with complex road network. This information is essential to be used as an input to air pollution models and further population exposure studies. Finally, quantification of the uncertainty range for the emissions opens a possibility to implement air pollution modelling for the study area using probabilistic approach.

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CHAPTER FIVE

5. MODELLING OF HUMAN EXPOSURE TO AIR POLLUTION IN THE URBAN ENVIRONMENT: A GPS BASED APPROACH

Submitted

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Abstract

The main objective of this work was the development of a new modelling tool for quantification of human exposure to traffic-related air pollution within distinct microenvironments by using a novel approach for trajectory analysis of the individuals. For this purpose, mobile phones with Global Positioning System technology have been used to collect daily trajectories of the individuals with higher temporal resolution and an algorithm based on trajectory data mining analysis was implemented within a Geographical Information System to obtain time-activity patterns. These data were combined with pollutants concentration fields provided by air pollution dispersion model. Additionally to outdoor, pollutant concentrations in distinct indoor microenvironments are characterised using a probabilistic approach. An example of the application for PM2.5 is presented and discussed. The results obtained for daily average individual exposure correspond to mean value of 10.6 $\mu g.m^{-3}$ and 6.0 – 16.4 $\mu g.m^{-3}$ in terms of 5^{th} – 95^{th} percentiles. Analysis of the results shows that using of the point air quality measurements for exposure assessment will not explain the individual variability. The methodology developed and implemented in this work provides time-sequence of the exposure events thus making possible association of the exposure with the individual activities and delivers main statistics on individual's air pollution exposure.

Keywords: exposure assessment, air pollution, traffic-related, GPS, GIS, trajectory data mining.

5.1. Introduction

Exposure to air pollution is estimated to cause 1.3 million deaths worldwide per year in urban areas and emissions from road traffic account for a significant share of this burden (WHO, 2011). In the last years, there has been an increase of scientific studies confirming that short and long-term exposure to traffic-related air pollutants leads to adverse health effects, including asthma, non-asthma respiratory symptoms, impaired lung function, cardiovascular mortality and morbidity (Brunekreef and Holgate, 2002; HEI, 2010). Therefore, an accurate assessment of human exposure is crucial for a correct determination of the association between the traffic-related air pollutants and the negative health outcomes (Hertel et al., 2008).

The assessment of exposure emerged as an important area of scientific research. Exposure estimates to atmospheric pollutants can address individuals (personal exposure) or large population groups (population exposure), and can be based on direct (exposure monitoring) or indirect methods (exposure modelling) (Zou et al., 2009). In practice, monitoring of personal exposure is limited to studies with a small number of individuals due to the high costs associated with the measurements. In the same sense, air quality time series provided by a monitoring network are frequently used as a good individual exposure indicator. Nevertheless, this estimate has been found to correlate poorly with personal exposures (Pellizzari et al., 1999; Oglesby et al., 2000; Koistinen et al., 2001; Kousa et al., 2002).

Several studies reveal that personal exposures tend to be greater in magnitude and more variable in location and time than the corresponding outdoor concentrations (Hatzopoulou and Miller, 2010). Individual exposure is then particularly sensitive to high spatial and temporal variations in outdoor concentrations and the "microenvironmental" variations imposed by a variety of indoor and outdoor locations (occupational, residential, etc.) (Georgopoulos et al., 2009). In this sense, outdoor concentration should not be used as an exposure indicator since it does not capture spatial heterogeneity in exposure to air pollution, time spent indoors and population mobility (Koistinen et al., 2001) thus leading to inaccuracies and underestimation of the effects of air pollution (Thomas et al.,1993; Szpiro et al., 2008;Peng and Bell, 2010). In addition, the presence of individuals in direct vicinity to the emission sources may results in higher exposure concentrations then pollution levels registered at monitoring stations (Baklanov et al., 2007). Therefore, combining air quality concentrations with time-activity patterns is crucial in assessing actual personal exposure to air pollution (Son et al., 2010).

In this perspective, exposure modelling technique arises as an alternative approach able to address the spatial and temporal variability of individual exposure concentrations and is recommended for exposure assessment (Schwela et al., 2002). Exposure modelling allows to determine exposures for individuals, defined population subgroups or entire populations, taking into account either real or hypothetical scenarios (Klepeis, 2006) and are typically used to supplement the monitoring data where direct measurement are not available. As results, exposure modelling can predict future exposures, as well as reconstruct historical exposure and the contribution of different chemicals can be clearly distinguished in exposure assessment (WHO, 2005b; Zou et al., 2009).

Exposure models constitute important tools providing quantitative evaluation of human exposure to environmental pollution, and its development has been identified as a priority area for future research (Brunekreef and Holgate, 2002; Brauer et al., 2002). Nowadays, a number of exposure models are available to support quantitative exposure analyses and assessments to air pollutants and, according to their characteristics and modelling procedures, they could be categorized as proximity models, interpolation models, land use regression models, dispersion models, integrated emission-meteorological models, and hybrid models (Jerret et al., 2005).

Air pollution exposure models can be developed to calculate short-term exposures (i.e. 1 hour or shorter in duration) or long-term exposures. Most of currently available exposure models have been designed to estimate human exposure to several regulated air pollutants (Johnson et al., 1999; Burke et al., 2001; Kruize et al., 2003), however a couple of models is also able to account for human exposure to hazardous air pollutants (MacIntosh et al., 1995; Özkaynak et al., 2008). They have been designed to quantify individual exposures as well as population exposures at the census level.

For exposure estimates outdoor pollution levels may be considered in combination with microenvironmental concentrations obtained from mass balance or empirical indoor/outdoor relationships (Georgopoulos et al., 2009). Additionally, population should be characterized by demographics and their time-activity patterns based on participant's diary or time–activity measurement databases (Burke et al., 2001; Kruize et al., 2003; Georgopoulos, 2005; Klepeis, 2006; USEPA, 2006a; USEPA, 2006b; Özkaynak et al., 2008; HEI, 2010). Extensive datasets on activity patterns and microenvironmental parameters are available for microenvironmental modelling (Freijer et al., 1998, McCurdy et al., 2000, Klepeis et al., 2001) providing additional information for probabilistic modelling and allowing an additional knowledge on the variability and uncertainty associated with exposure estimates (Zou et al., 2009). It is important to highlight that variability represents true heterogeneity, diversity, inter-individual differences, temporal changes, etc. in an input parameter while uncertainty reflects a lack of knowledge of the true value (Frey, 1992; Hertwich et al., 2000; WHO, 2000). Parameter variability and uncertainty represent the sources of uncertainty that have received most attention in human exposure modelling (Fryer et al., 2006). In addition, recent advances have also occurred in the development of GIS-based exposure models, which attempt to reproduce the spatial and temporal dynamics of air pollution and population mobility (Gulliver and Briggs, 2005; Zhan et al., 2006; Wheeler et al., 2008).

However, information on the actual "activity space" of individuals required for high resolution exposure modelling is rarely available, and home addresses are generally used as the surrogate for the personal exposure, when in fact a high percentage of an individual's exposure can accrue from relatively short periods of time spent in high-polluted microenvironments (HEI, 2010). In this perspective, the time-sequence of exposure events is not preserved in exposure assessment, and the information to evaluate possible correlations in exposures to different pollutants due to activities that are related in time is not conserved. The source-receptor relationship, especially for "hot-spots" peak exposure is still insufficiently addressed and the contribution of traffic- related air pollution to the total exposure is not clear (Wang et al., 2009; HEI, 2010). In addition, the development of innovative models that reduce uncertainties in exposure characterization is required (Lioy, 2010). Furthermore, the relationship between the exposure concentration, which vary substantially with geographical location, and the exposure duration, which is related with human activities, is still insufficiently addressed. Recent findings highlights that the population mobility is one of the factors that may affect significantly the exposure (Nethery et al., 2008; Beckx et al., 2009; Dons et al., 2011; Tchepel and Dias, 2011).

In this sense, the knowledge of where individuals spend time is essential for assessment of human exposure to air pollution and research on human behaviour or activities is a crucial component of modern and future exposure science (Lioy, 2010). To address this issue, the availability of enhanced resources such as geographic information system (GIS), global positioning system (GPS) and data mining techniques, could be used to analyse the human behaviours and activities required for exposure assessment, opening new perspectives to quantify human exposure to traffic-related air pollution.

One of the problems of the exposure assessment approaches is the uncertainty related with human mobility during the exposure assessment period. Predictability in human dynamics by studying the mobility patterns of individuals using mobile phones became an emerging field (Song et al., 2010) and GPS technology presents as a promising tool by monitoring real-time geographic positions. GPS-equipped mobile phones can record the latitude-longitude position of individuals at each moment, offering many advantages over traditional time-location analysis, such as high temporal resolution, and minimum reporting burden for participants (Rainham et al., 2010).

The GPS technology guarantees that there will be an increasing availability of large amounts of data affecting to individual trajectories, at increasing localization precision. However, there is a challenge to extract, the spatio-temporal patterns from these trajectories that convey useful knowledge (Zheng and Zhou, 2011). Thus, the data mining appears as a validated technique to automatically identify time-activity location in major microenvironments, such as commuting, indoor, and outdoor locations (Wu et al., 2010). Data mining is used to search through large amount of raw data in order to find useful data. The goal of this technique is to identify relevant and important patterns that were previously unknown (Larose, 2006; Witten and Frank, 2005).

The present work intends to develop a new modelling tool for quantification of human exposure to traffic-related air pollutants by using a novel approach based on trajectory analysis of individuals and air pollution modelling with high spatial-temporal resolution. For this purpose, information on pollutant concentrations at different microenvironments and detailed time-location data collected for each individual by mobile phones with GPS are processed using trajectory data mining and geo-spatial analysis within GIS. Also, the model integrates a probabilistic approach to estimate the variability of the microenvironmental parameters in the predicted individual exposure. The development of a GPS based EXPOSure model to traffIc-relaTed aIr pOllutioN (ExPOSITION) is presented and described.

5.2. Methodology - Human exposure modelling

The ExPOSITION model is developed to assess average short (e.g. daily) and long-term (e.g. annual) inhalation exposures of the individuals to traffic-related air pollutants over urban spatial scale with high spatial-temporal resolution. For this purpose, air pollution concentrations are estimated for different microenvironments (described in Section 5.2.1) and combined with detailed time-activity patterns obtained from data collected by mobile phones with GPS technology (described in Section 5.2.2 and Section 5.2.3). The ExPOSITION modelling system developed and applied in this study is schematically presented in Figure 5.1 and described in the following sections.

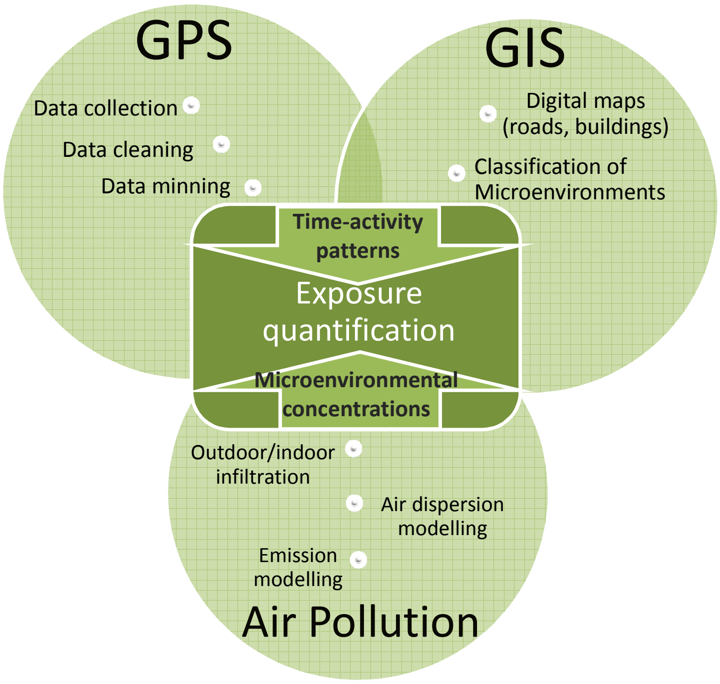

Figure 5.1. Conceptual framework of the ExPOSITION modelling system.

The time-activity patterns are determined by the model based on a novel approach developed for collection and analysis of data registered by mobile phones with GPS technology and thus providing the daily trajectories of individuals required for the exposure assessment. A GPS mobile phone combined with a GPS tracking software is used to determine the precise location of a person and to record the position at regular time intervals. Time-location information is obtained from geographic coordinates, speed and time recorded and stored by the GPS tracking system that characterise the movements of individuals in time and space during their daily activities. To process the GPS data an algorithm based on trajectory data mining has been developed and an algorithm for classification of microenvironments has been implemented within GIS.

Personal exposure is characterised by ExPOSITION model in terms of time-weighted average exposure concentration calculated from air pollutant concentration fields and time spent by individuals in different microenvironments (Equation 5.1). It is important to highlight the distinction between air pollution “concentration” provided by dispersion models, and “exposure concentration” defined as amount of chemicals that comes into contact with the human body and take into account not only pollutant concentration fields but also the location of an individual and duration of the exposure. Thus, individual exposure is calculated by ExPOSITION as following:

$$\overline{E}_i = \frac{1}{t_2 - t_1} \int_{t_1}^{t_2} C(x,y,z,t)_i \, dt \qquad (5.1)$$

where $\overline{E}_i$ (µg.m^{-3}) is the average exposure concentration for person i, $C(x,y,z,t)_i$ (µg.m^{-3}) is the air pollutant concentration occurring at a particular point where the person i is located during the time t and spatial coordinate (x,y,z) and t_1 and t_2 (h) are the starting and ending times of the exposure event.

Exposure estimates are provided in µg.m^{-3} and can be determined for each individual as hourly, daily or annual average and resulting data can be exported for further analysis (e.g. epidemiological analysis and health impact assessment).

5.2.1. Microenvironmental concentrations

Specific microenvironments are distinguished in the exposure model including residence, other indoors, outdoors, and in-vehicle (Table 5.1). Two different approaches are considered to characterise pollution levels in these microenvironments. Thus, outdoor concentrations are estimated using atmospheric dispersion modelling and different modelling tools may be used to provide this external information for ExPOSITION as will be discussed in section 5.3. For indoors and in-vehicle microenvironments a probabilistic approach was implemented as an integrated part of ExPOSITION algorithm. In this case it is assumed that within a microenvironment the pollutants are homogeneously distributed and microenvironmental concentration $C(x,y,z,t)$ (µg.m^{-3}) considered in Equation 5.1 is calculated using a linear regression equation based on the outdoor/indoor infiltration factor α_j (dimensionless) and additional contribution of indoor pollution sources expressed as β_j (µg.m^{-3}):

$$C(x,y,z,t) = \beta_j + \alpha_j \times C(x,y,z,t)_{ambient} \qquad (5.2)$$

where $C(x,y,z,t)_{ambient}$ (µg.m^{-3}) is the outdoor concentration that occurring in the immediate vicinity to the microenvironment j at time t and spatial coordinate (x,y,z).

Microenvironmental concentrations are estimated based on a probabilistic approach considered by the model that attempts to capture the variability in microenvironment parameters. In this sense, to calculate microenvironmental concentrations for each individual the ExPOSITION model randomly assigns the parameters β and α to each indoor location from empirical distributions taking into account the average and standard deviation obtained from literature review for each type of microenvironments (Table 5.1).

Table 5.1. Parameters used to determine PM2.5 concentrations in different microenvironments.

Microenvironment	β		α		Data source
	average	standard deviation	average	standard deviation	
Residence	5.75	3.91	0.41	0.06	Hoek et al., 2008
Vehicle (no smoking)	33.00	7.20	0.26	0.14	Burke et al., 2001
Office (no smoking)	3.60	1.30	0.18	0.06	
School	6.80	1.40	0.60	0.09	
Public access	9.00	3.60	0.74	0.18	
Restaurant/Bar	9.80	0.50	1.00	0.05	

A single value is selected from the probabilistic distribution of each microenvironmental parameter α and β. These values are then used in the model to produce a single estimate of microenvironmental concentration. This process is repeated many times, with new values for each stochastic input parameter and probability distribution of exposure in the microenvironments is obtained.

5.2.2. Trajectory data mining

Trajectories of the individuals are required as one of the main inputs to the exposure modelling. Collection of time-location information using GPS technology provides continuous tracking of the individuals with high data resolution in time and in space. However, significant uncertainties associated with the processing and classifying of raw GPS data is one of challenging issue for the exposure studies (USEPA, 1992; Wu et al., 2010). To overcome some of the limitations, automatic processing of GPS raw data using the trajectory data mining is implemented in ExPOSITION model.

In order to identify important patterns, several levels of GPS data processing are required (Figure 5.2). First, it is necessary to “clean” the GPS raw data to eliminate invalid entries. At next, the places where the individual was stopped for a certain time period are distinguished from moving activities, like driving a vehicle. And finally, it is necessary to discover which of these points belong to the same activity/place. For this purpose the data clustering process is implemented to distinguish significant places based on the analysis of spatial and temporal information of GPS points (Figure 5.2).

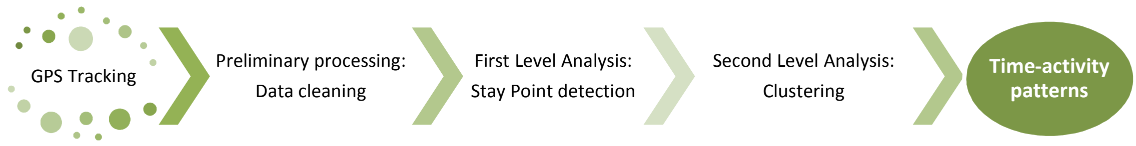

Figure 5.2. Schematic representation of the trajectory data mining analysis.

Thus, “significant places” are considered as those locations that play significant role in the activities of a person, carrying a particular semantic meaning such as the living and working places, the restaurant and shopping mall, etc., ignoring the transition between these places. Additionally, a “movement activity” is a composition of movements with a frequent regularity of location change over time which can be aggregated by the purpose of the trip of an individual.

A preliminary processing of GPS data is implemented as a first step to “clean” the data and converts it into a standard format in preparation for the clustering approach. For this purpose an error-checking algorithm was developed to remove invalid points. This algorithm considers a measurement as valid, if the GPS receiver is able to see at least four satellites and if the horizontal dilution of precision (HDOP) value is below 6 (Figure 5.3). Otherwise the measurement is considered invalid. Also, the algorithm evaluates incorrect entries of the travel speed.

GPS datasets provide information on the locations in coordinate form (e.g. latitude and longitude) but contains no semantic meaning (Zhou et al., 2007a) like the address or characteristics of location, i.e. type of microenvironments. Therefore, it is necessary to extract and distinguish in the GPS data the locations where the individual stopped for a certain time period and these locations are designated as “stay points”. A stay point represents a geographic location in which the individual stays for a certain time period and in addition to a raw GPS point carries a particular semantic meaning.

The algorithm to extract stay points from GPS data is iterative and it is based on searching for locations where the user has spent a longer time period (Li et al., 2008). As presented in Figure 5.3, the extraction of a stay point S from a user’s GPS trajectory $P = \{p_1, p_2, \dots, p_K\}$, depends on two scale parameters: a distance threshold (D_{threh}) and a time threshold (T_{threh}). Thus, a single stay point S can be characterized by a group of consecutive GPS points p_i containing latitude ($p_i.Lat$), longitude ($p_i.Long$) and time ($p_i.T$):

$S = \{p_i\}$, where $m \leq i \leq n$,

$Distance(pm,p_i) \leq D_{threh}$ and

$|p_n.T - p_m.T| \geq T_{threh}$

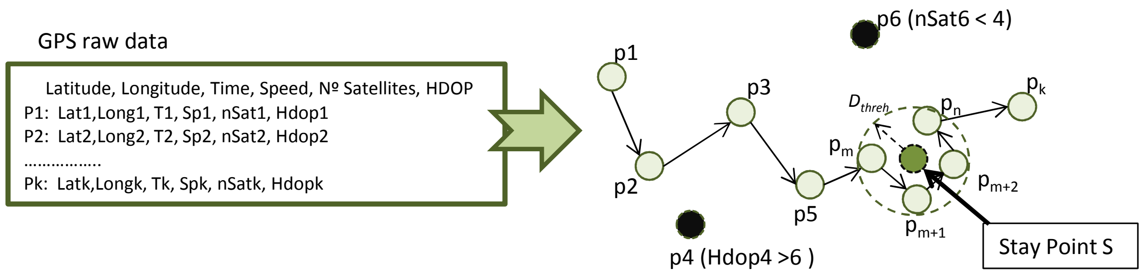

Figure 5.3. GPS raw data, GPS “clean” trajectory and stay points detection.

A pre-processing of the GPS data and detection of the stay points is important to extract some important locations. However, the repetition of the same locations is not considered and each time that a location is discovered it is assumed as a new location. To overcome this problem a second level analysis to group up different stay points with the same semantic meaning is implemented using cluster analysis.

Clustering is a data mining technique focused on detecting hidden groups, or clusters, among a set of objects (Bock, 1996). In this study, in order to group the points belonging to the same premises, and thus define the personally significant places, a density-based clustering algorithm DJ-Cluster (Zhou et al., 2004; 2007a; 2007b) was implemented. The DJ-Cluster algorithm is selected and applied in this study, since it is less vulnerable to noise and does not require the number of places as a parameter. However, the algorithm depends excessively on the density of the points and does not give importance to the time spent in each site, i.e. duration, which will be relevant for the exposure quantification.

In the clustering algorithm, the neighbourhoods within distance Eps are analysed for each point. If at least a minimum number ($MinPts$) of such neighbourhoods is found, the points are either grouped as a new cluster or joined with an existing cluster, and a significant place is created. Otherwise, the point is labelled as a moving activity (e.g. being in vehicle microenvironment) (Figure 5.4). The following conditions define the density-based neighbourhood of a point and density-joinable relationships (Zhou et al., 2007a):

a) Density-based neighbourhood of a point:

The density-based neighbourhood N of a point p, denoted by N(p), is defined as:

$$N(p) = \{q \in Q | dist(p,q) \leq Eps\} \tag{5.3}$$

where Q is the set of all points, q is any point in the sample, Eps is the radius of a circle around p to defines the density. The following condition is also needs to be satisfied for $N(p)$:

$$\# N(p) >= MinPts \qquad (5.\ 4)$$

where $MinPts$ is the minimum number of points required in that circle.

b) Density-Joinable:

$N(p)$ is density-joinable to $N(q)$ denoted as $J(N(p),N(q))$, with respect to Eps and $MinPts$, if there is a point such that both $N(p)$ and $N(q)$ contain it.

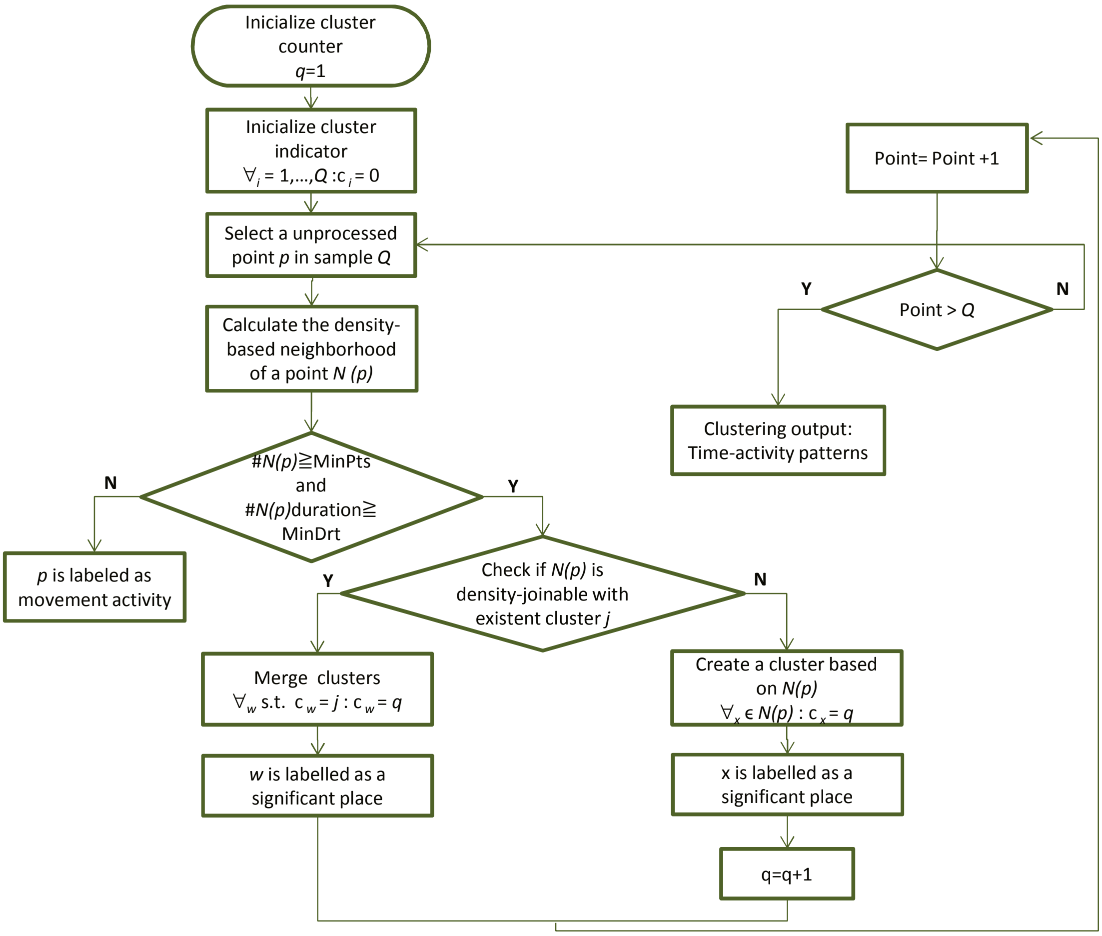

Figure 5.4. Flowchart of the clustering process.

For the objectives of this study the sites are identified as personally significant places talking into account two variables: density and duration. In this perspective, DJ-Cluster algorithm was changed in order to implement additional condition based on duration of stay, as presented in Figure 5.4. Thus, $N(p)$ defined in Equation 5.3 needs to satisfy simultaneously two conditions:

$$\# N(p) >= MinPts \wedge N(p).duration >= MinDrt \tag{5.5}$$

where $MinDrt$ is a parameter that represents the minimum duration at a location. Thus, p can be considered as a cluster or merged with an existent cluster in case that has a minimum number of points required $MinPts$ and a minimum duration $MinDrt$.

These data are further analysed within GIS environment for classification of microenvironments and to obtain information on time-activity patterns.

5.2.3. Time-activity patterns

Location of the individuals in space and in time is required to estimate individual exposure in a combination with pollutants concentration fields provided by air pollution dispersion model.

In order to obtain information on time-activity patterns the significant places and movement activities extracted from the trajectory are further analysed within GIS environment in order to cross this information with other geo-spatial information. For this purpose, geoprocessing of GPS data is performed using ModelBuilder module provided by ArcGis 10. ModelBuilder can be thought of as a visual programming language for building workflows in which it is possible to create, edit, and manage geospatial analysis (Allen, 2011).

The geoprocessing of GPS data is accomplished by considering analytical functions and several predefined criteria based on speed, time and spatial location register for the trajectory points to classify the significant places and movement activities to three activity categories: indoor, outdoor and in vehicle travel. The detailed GIS-maps are used to identify and to classify the microenvironments.

An indoor activity is distinguished from outdoor based on the time register. If the spending time in that point is equal or higher than 10 minutes, based on several tests carried out in this study and as presented by Ashbrook and Starner (2003), the significant place is identified as an indoor activity, and it is geographically located to the nearest indoor microenvironment, acquiring the entire attribute data associated to this microenvironment, such as microenvironment type (residence, workplace, restaurant, etc.). Additionally, the speed value is analysed in order to distinguish outdoor activity from in vehicle travel. However, the higher speed values registered during driving a vehicle are not sufficient to identify a movement activity. Also, activities like being static outdoor and in the traffic jam are difficult to distinguish based on speed criteria only. Thus, if the speed value is less than the speed of walking of 2 km.h^{-1} (TRB, 1994) the distance between the identified point and the nearest road will be analysed. If there is no intersection with the road network, the significant place is identified as an outdoor microenvironment, such as being in a park, sitting on a terrace, etc. Otherwise, vehicle microenvironment is identified.

Finally, this detailed time-activity patterns for each individual will be linked with the pollutants concentration fields varying in space and in time provided by air pollution dispersion model described in the next section, allowing to produce exposure estimates within distinct microenvironments.

5.3. Emission and Air quality modelling

Air quality modelling allows establishing the relationships between current emissions and current air quality at particular locations. Information on variability of air pollutant concentrations is essential for the exposure quantification and these data may be provided for ExPOSITION by any modelling tools if it is compatible with their requirements in terms of spatial and temporal data resolution.

In this work, hourly traffic emissions required by the air quality model were estimated using the Transport Emission Model for Line Sources (TREM). The emission factors considered by TREM depend on average speed, fuel type, engine capacity and emission reduction technology. A new version TREM-HAP (Transport Emission Model for Hazardous Air Pollutants) prepared to calculate HAPs emissions (Tchepel et al., 2012) has been used to provide inputs for AUSTAL2000 dispersion model.

AUSTAL2000 is the official reference air dispersion model of the German Regulation on Air Quality Control for short-range applications (Janicke and Janicke, 2002; Janicke, 2004). The model is based on Lagrangian approach that simulates the dispersion of air pollutants by utilizing a random walk process. Three-dimensional diagnostic wind fields is calculated based on a given initial wind profile and a given terrain profile and/or set of building shapes. Additionally, the vector of the turbulent velocity is randomly varied for every particle by using a Markov process (Janicke, 2002; VDI, 2000). The fundamental equation for the Lagrangian atmospheric dispersion of a single pollutant is given by Equation 5.6.

$$C(x,t) = \int_0^t \int P\left(x,t \mid x',t'\right) S\left(x',t'\right) dx' dt' \qquad (5.6)$$

where $C(x,t)$ is the average pollutant concentration in x at time t, $S(x',t')$ is the source term and $P(x,t \mid x',t')$ is the probability density function (PDF), that the hypothetical parcel moves from the point x'at time t' to the point x at time t. Therefore, if actual paths of the portions of air can be obtained, the simple calculation of the density of trajectories points provides an estimate of the concentration (Graff, 2002).

The main objective of AUSTAL2000 application in the current study is the calculation of atmospheric dispersion of substances, including PM fractions (4 different classes of the aerodynamic diameter) allowing to establish relationship between emissions and air quality, and to provide hourly pollutants concentration fields. Additionally to input data on emissions, a continuous time series of meteorological parameters, including wind direction, wind speed and atmospheric stability are required by AUSTAL2000.

Currently, several studies using the AUSTAL2000 are available, as well comparative analyses with other dispersion models (Yau et al., 2010; Langner et al., 2011; Merbitz et al., 2012; Gerharz and Pebesma, 2012).

5.4. Model application

The methodology was applied to Leiria urban area situated in the central part of Portugal and covering 8 sub-municipality units. The study domain covering an area of 4.5 x 4.5 km^2 with 20m grid resolution and a complex terrain, containing about 5000 buildings considered as obstacles for the air dispersion modelling. The Leiria urban area and road network considered in this study for the exposure quantification are presented in Figure 5.5.

Hourly PM2.5 emissions from road traffic were estimated by TREM based on the traffic volume for each road. For this purpose, data reported by Pinto et al. (2008) were used to characterise the number of vehicles for each road link. To estimate PM2.5 concentrations hourly simulations were conducted with AUSTAL2000 model taking into account hourly meteorological conditions and background concentrations given by the nearest background air quality monitoring station.

In order to characterize the variability in input parameters used to calculate microenvironmental concentrations (Equation 5.2), a set of random inputs characterizing the infiltration factor α and the contribution of indoor pollution β are generated for each microenvironment. The PDF for both parameters is determined using the information presented in Table 5.1. A combination of random values is used to create 625 independent inputs for each microenvironment to be considered by ExPOSITION for the exposure estimations.

TTGPSLogger tracking system (TTGPSLogger, 2012) was used to collect GPS data providing trajectories of 5 individuals during a working day of November 2010. TTGPSLogger is a GPS logger software for Symbian S60 allowing to store detailed time-location information on geographic coordinates, speed and time during its use over the daily activities of individuals. In addition, information on the positioning accuracy of GPS receiver is provided (number of satellites, position dilution of precision (PDOP), etc.). The GPS tracking log can be written in NMEA, GPX, or KML format. For proper implementation of the trajectory data mining analysis, the GPS data was collected in one-second intervals.

5.5. Results and Discussion

In this section, the results obtained with newly developed modelling tool for short-term PM2.5 exposure quantification are presented and discussed.

TTGPSLogger tracking system installed on mobile phone is used to collect real-time latitude-longitude position of individuals, speed and time during their daily activities (Figure 5.5a). This information was stored in a GPX file format that is compatible with GIS systems presenting very useful to analyse the spatial distribution of large amount of GPS raw data collected. Thus, during a typical working day of one of the individuals analysed in this study, 30179 GPS raw points with a temporal resolution of 1 second are collected by TTGPSLogger tracking system. However, some of the collected GPS data points with invalid information, such as incorrect entries of speed values achieving maximum of 650 $km.h^{-1}$, are identified.

Most of the invalid measurements observed in this study are from areas where the individual has stayed indoors due to the obstruction of the GPS signal inside of buildings. Furthermore, there are some situations where the GPS receiver located inside buildings does not lose the signal but the data collected are affected by significant errors achieving about 60 meters of distance from the actual position. Another limitation observed is a gap of GPS information during some periods (from 15 seconds to 10 minutes) depending on the GPS status.

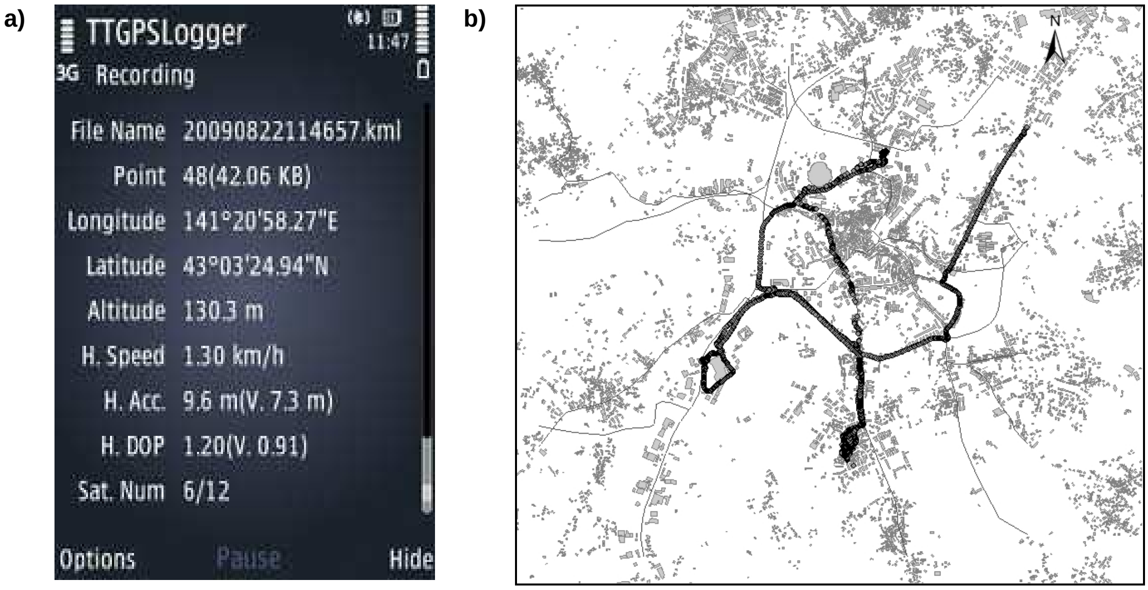

Figure 5.5. a) Data recording screen from mobile phone; b) Spatial visualization of the GPS raw data recorded.

Taking into account the limitations detected during the analysis of GPS raw data, cleaning of the data and their processing are required in order to predict the time-activity patterns (Figure 5.6).

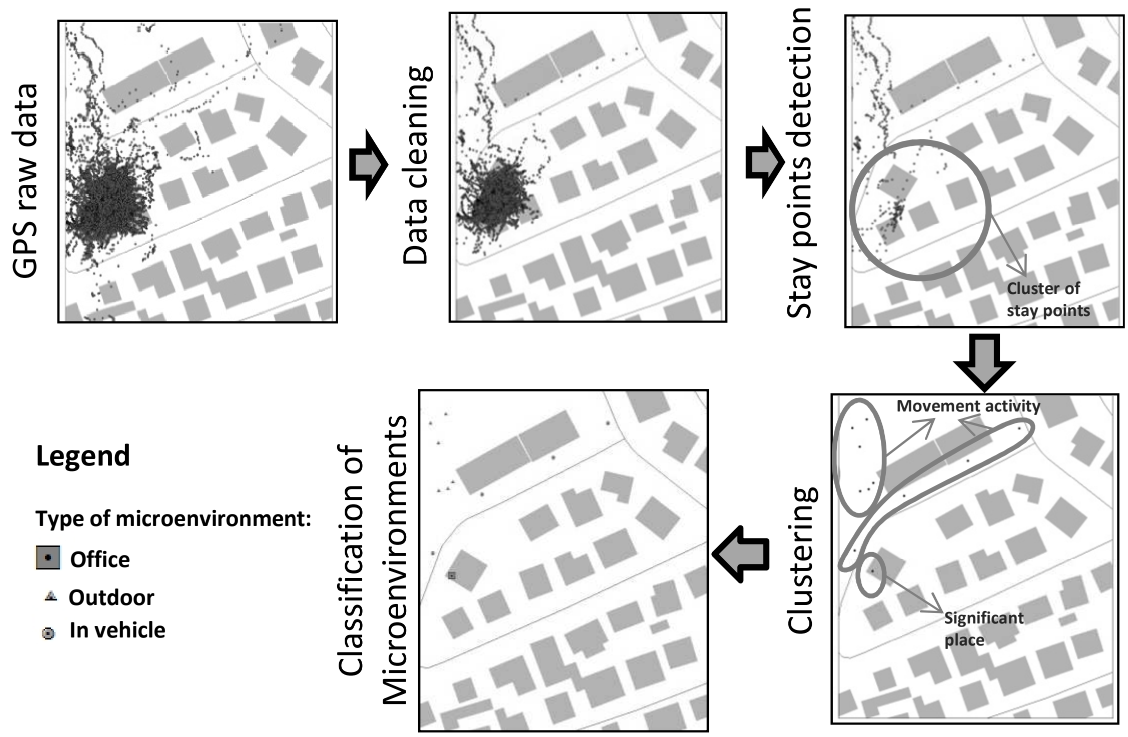

Figure 5.6. Example illustrating the data processing applied to GPS raw data.

In Figure 5.6 a sequence of images with a zoom to the workplace of the individual 4 is presented to illustrate the formation of 3 clusters detected from the data. The first image corresponds to the raw GPS points recorded. At next, the data cleaning allows to remove invalid points from collected GPS raw points. However, this approach is only a pre-processing to detect errors and inconsistencies in data. The stay point detection algorithm reduces significantly the number of GPS points that are consequently used for the clustering. In the example presented in Figure 5.6, one significant place and two movement activity clusters are identified from the set of stay points.

The locations resulting from the clustering algorithm are further analysed within GIS environment in order to cross this information with other geo-spatial information and to obtain detailed time-activity patterns classified by different types of microenvironments. Thus, in case of the individual 4, 30179 collected GPS raw points resulted in 15978 stay points, originating 295 locations that are linked with the pollutants concentration in distinct microenvironments to assess its individual exposure.

In order to estimate human exposure to PM2.5, hourly traffic emissions and air pollutants concentrations were estimated. Figure 5.7a illustrates the spatial variations in hourly traffic-related emissions across the study area obtained by linking TREM-HAP outputs to GIS maps. As could be seen in the figure, higher emission values are observed for main city entrances.

a) **b)**

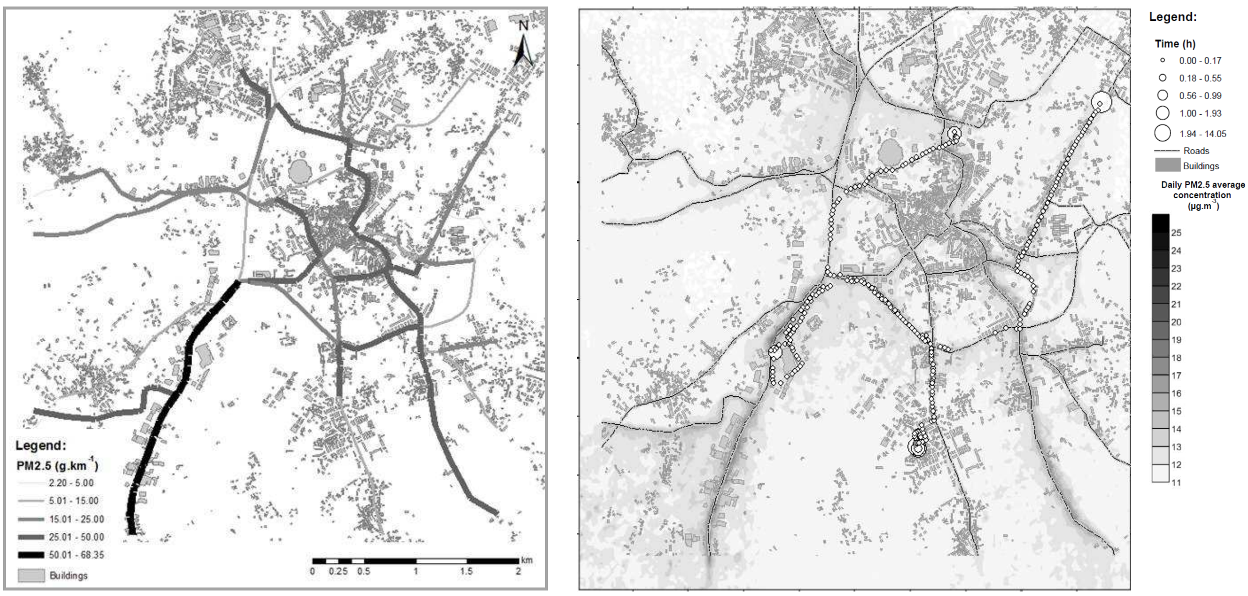

Figure 5.7. Spatial distribution of a) hourly PM2.5 emissions (g.km^{-1}) and b) daily average PM2.5 concentration (µg.m^{-3}) and time spent by the individual in each microenvironment.

The spatial distribution of the air pollutants concentration obtained by the AUSTAL2000 is presented in Figure 5.7b showing that distribution of pollution levels within the study domain is not homogeneous. Also, time-activity patterns obtained for one of the individuals are presented in the figure as an example. The analysis of results examines the PM2.5 concentration variation in space and in time provided by air pollution dispersion model and the influence of time spent in each microenvironment type. Thus these findings enhance the importance of taking into account the high spatial and temporal variations in outdoor concentrations, the "microenvironmental" variations imposed by a variety of indoor and outdoor locations and the time spent indoors to obtain accurate personal exposure estimates to air pollution.

For better understanding of the contribution of different microenvironments to the daily average PM2.5 exposure in the study area at a typical working day, several statistical parameters, including average individual exposure, 5th and 95th percentile and extreme values were analysed (Table 5.2).

Table 5.2. Exposure concentration for PM2.5 ($\mu g.m^{-3}$) in different microenvironments.

Microenvironment	Average	Percentile 5	Percentile 95	Minimum	Maximum
Residence	10.2	7.7	17.8	7.0	18.1
Workplace	8.7	4.5	11.7	4.5	16.1
Public Access	14.5	13.5	16.3	13.5	26.1
Bar/Restaurant	16.7	15.2	17.9	15.0	18.0
Vehicle	35.2	34.6	37.4	20.2	44.6
Outdoor	7.5	5.2	12.7	4.8	41.6

As could be seen in Table 5.2, the largest variability in the exposure concentration is identified for outdoor and residence microenvironment. Exposure concentration calculated for in vehicle are characterised by smaller variability range but higher absolute values in comparison with the other types of microenvironments. In addition, it is possible to verify that the variability in the PM2.5 exposure concentration in each microenvironment type is significant showing the importance to consider this variability in individual exposure modelling.

As expected, the indoor microenvironments represent a great relevance for the exposure of individuals (Figure 5.8). On the other hand, it is possible to verify that being outdoors represents a very low contribution to the exposure because corresponds only about 2% of the time spent by individuals during their daily activities, which suggests that outdoor concentrations measurements should be used carefully for human exposure quantification. However, outdoor concentrations represent an important part of the pollution levels estimated for indoor microenvironments due to outdoor/indoor infiltration.

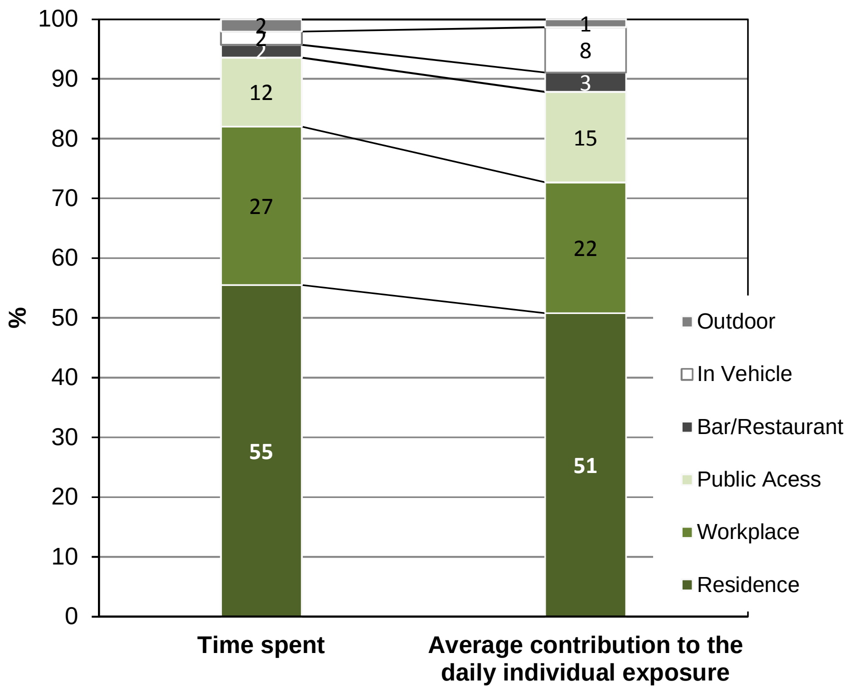

Figure 5.8. Distribution of time spent by individuals and average contribution of different microenvironments to daily individual exposure.

In order to better understand the individual exposure obtained during the simulation period, a temporal variation of the exposure concentration was analysed as presented in Figure 5.9. Several statistical parameters, including hourly average exposure concentration, 5^{th} and 95^{th} percentile are analysed for each individual.

The results show that the 5 individuals are exposed to different PM2.5 concentrations during their daily activities, and a significant variability in PM2.5 exposures across the individuals is evident in Figure 5.9. Analysing the individual exposure concentrations during night time (until 7:00 (7a.m.) approximately), when the people stay in residence, the hourly exposure concentrations presents a similar trend with the outdoor concentrations but different magnitude. However, throughout the day and depending on the daily activity of the individuals the hourly average exposure concentrations tend to be more variable. The highest exposure levels are related with both the magnitude of pollutant concentrations and the time spent in specific microenvironments as, for example, could be seen in Figure 5.9 for the individual 1 at 16:00 (4 p.m.).

Overall, the daily average exposure to PM2.5 predicted by the ExPOSITION model correspond to 10.6 $\mu g.m^{-3}$ in terms of the mean value for all individuals and 6.0 – 16.4

μg.m^{-3} in terms of 5th – 95th percentiles. Comparing the mean value obtained by the model and estimated from air quality measurements at a fixed point (11 μg.m^{-3}), an agreement between the approaches was evidenced. However, the ExPOSITION model reveals additional inter and intra-variability of individuals' exposure levels, suggesting limited representativeness of air quality concentrations obtained from point measurements to characterize individual exposure to urban air pollution.

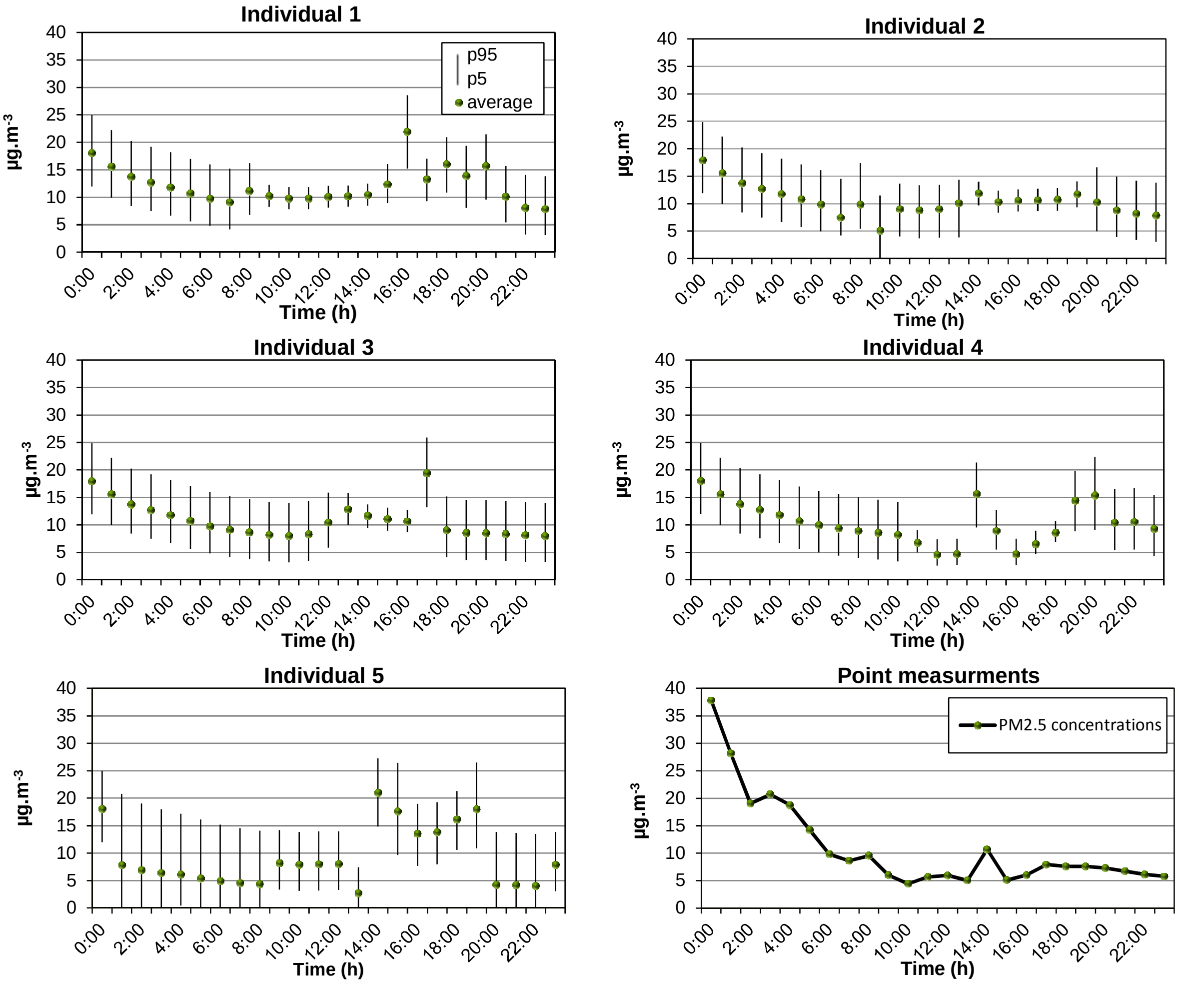

Figure 5.9. Temporal variation of individual exposure concentrations (average, 5th percentile and 95th percentile) and outdoor concentrations of PM2.5.

The results obtained from the ExPOSITION model are in good agreement with the daily average exposure reported for other European cities such as Helsinki (9.9 μg.m^{-3}) (Koistinen et al., 2001) and Amsterdam (14.5 μg.m^{-3}) (Janssen et al., 2005). The current study shows that high PM2.5 exposure is mainly attributed to indoor microenvironments rather than outdoor, as also presented by Georgopoulos (2005). In this context individual time-activities patterns and time spent at different microenvironments during the day should be of prime concern additionally to the variability in the pollution levels, as presented by Burke et al. (2001).

5.6. Conclusions

A GIS-based human exposure model able to estimate the individual exposure to traffic-related air pollutants with high spatial-temporal resolution has been developed and implemented using advanced GIS tools and GPS tracking system. The current work provides a description of the methodology and an application example to characterise the individual exposure at the spatial and temporal scales defined by the microenvironments and exposure activity events by using a novel approach for trajectory analysis of the individuals based on a mobile phone GPS tracking system.

Under this work, a time-activity pattern discovery sequence, based on trajectory data mining and geo-spatial analysis within GIS, was developed to extract useful time-location information from GPS raw data collected by a mobile phone with a GPS tracking system carried by the user during their daily activities. Taking into account the limitations detected during the analysis of GPS raw data, the results obtained during the several levels of GPS data analyses indicate that this approach could be used to analyse the human behaviours and activities required for exposure assessment.

Time series of individual exposure concentrations to PM2.5 are presented for the entire study area characterizing a person’s contact with a given pollution levels at different microenvironments. The results show a significant contribution of indoor microenvironments to the total exposure values thus stressing that individual exposure depends not only on the exposure pollution levels but also on the time spent in the microenvironment during the day.

The methodology developed and applied in this study preserves time-sequence of the exposure events thus making possible association between the exposure and individual activities, providing thus information on individual exposure taking into account where individuals spend their time and the high spatial and temporal variations of the “microenvironmental" concentrations imposed by a variety of indoor and outdoor locations.

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CHAPTER SIX

6. MODELLING OF HUMAN EXPOSURE TO BENZENE IN URBAN ENVIRONMENTS

Submitted

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Abstract

Urban areas characterized by high spatial and temporal variability in air pollution levels require implementation of comprehensive approaches to address exposure of individuals. The main objective of this work is to implement a quantitative assessment of the individual exposure to benzene in urban environment. For this purpose, the ExPOSITION model based on GPS-tracking approach is applied to estimate individual exposure in different microenvironments. The current work provides an application example and validation of the modelling approach against personal and biological exposure measurements collected during the measurements campaign. The results obtained for daily average individual exposure to benzene correspond to mean value of 1.6 $\mu g.m^{-3}$ and 0.8 to 2.7 $\mu g.m^{-3}$ in terms of 5^{th} to 95^{th} percentiles. Validation of the model results against several personal exposure samples collected for the selected individuals reveal a Pearson's correlation coefficient of 0.66 ($P<0.0001$, 95% CI 0.42 to 0.82). The modelling approach presented in this work explicitly addresses the temporal and spatial variability in the exposure and establishes source-receptor relationship, thus providing more consistent results in comparison with the personal exposure estimates based on home address outdoor concentrations.

Keywords: Exposure assessment, benzene, personal exposure monitoring, biomonitoring, exposure model validation.

6.1. Introduction

Among the extended number of chemicals emitted by road traffic sources, hazardous air pollutants (HAPs) require special attention due to growing international recognition of their link with a variety of adverse effects on human health and the need for action to minimize these risks (HEI, 2010). One of the HAPs of prime concern to human health is benzene, defined as one of the most important health-based European Union priority substances (Bruinen de Bruin et al., 2008).

The main source of benzene emissions in urban areas is road transport (Johnson et al., 2007; Weisel, 2010), contributing about 85% for outdoor benzene levels (EEA, 2007). In this concern, characterization of the transport activity and the quantification of corresponding emissions in urban areas where inhabitants are living close to the pollution sources are required for better human exposure analysis. For this purpose, transportation modelling linked with the emissions and dispersion models is considered as one of more suitable approaches to provide detailed information concerning traffic flux for each road segment and related pollution (Borrego et al., 2006). Additionally to on-road vehicle exhaust emissions, the exposures to outdoor benzene are likely to occur during the refuelling at fuel stations and near gasoline fuel stations (Weisel, 2010; VANR, 2011) which will vary according to content of fuel, the presence or absence of vapour control devices and the amount of time spent at such locations (Duarte-Davidson et al., 2001).

The growing concern about adverse health effects of exposure to benzene related even with typical ambient concentrations led to the need for monitoring of its outdoor concentrations as well as non-occupational personal exposure of several population groups (Cocheo et al., 2000; Tchepel et al., 2007; Weisel, 2010). Several studies have reported that daily mean ambient air concentrations of benzene in rural areas are in the range of approximately 0.7 – 1 $\mu g.m^{-3}$, but in urban areas the concentrations are reported in the range of 1.6 – 20 µg.m-3 (WHO, 2000; HEI, 2010). Higher values have been measured in some cities with high traffic density and unfavourable meteorological or geographical conditions (WHO, 2000; Deole et al., 2004; Farmer et al., 2005). Currently, in order to avoid, prevent or reduce harmful effects on human health and the environment as a whole, European Directive 2000/69/EC establishes 5 $\mu g.m^{-3}$ (calendar year or annual mean) as the limit value for benzene concentration in ambient air.

The contribution of indoor microenvironments, where people spend 80 to 93% of their time, to the individual benzene exposure has been increasingly recognized as being of importance (Klepeis et al., 2001; Adgate et al., 2004; Phillips et al., 2005). Additionally to infiltration of outdoor air pollution, a variety of substantial indoor sources of benzene, such as tobacco smoke, usage of petroleum-related fuels for cooking/heating and benzene emitting cleaning/consumer products may contribute to the individual exposure. Also, several findings indicate that indoor concentrations of benzene are typically higher than the respective ambient levels (George et al., 2011). However, despite the research community recognizing the importance of indoor environments in personal exposure, non-occupational air pollution regulations have typically been applied focusing on outdoor rather than indoor air. For this reason, the amounts of air pollutants to which general populations are actually exposed are rarely quantified (HEI, 2010).

Under this context, individual exposure modelling technique are arising as an alternative and effective approach able to address the spatial and temporal variability of individual exposure (Setton et al., 2011; Steinle et al., 2013). Although previous studies have analysed the distribution of concentrations and much work has been conducted toward modelling population exposures to air pollutants using information collected in time/activity diaries and microenvironment concentrations, very little has been done toward validating of such models at the level of the individual. Assessing the validity of the exposure estimates from models is often not straight forward, but it is essential for the credibility of the models. Therefore, the validation of models with independent data sets (e.g. from biomonitoring and personal exposure monitoring) is useful to check whether the proposed models serve as surrogates for individual exposure and to know the extent of the exposure estimation error, which should be accounted for in epidemiologic studies and risk assessments (Fryer et al., 2006; Liu et al., 2007). Personal monitoring may be performed with active monitors or passive samplers, and is considered the most accurate estimate of a person’s ‘true’ exposure and the mobility of people across various microenvironments, according to their daily activities (Carrer et al., 2000). However, some studies reveal that its wide-scale application to evaluate exposures at the population level is limited due to their cost and sometimes even impractical for certain subpopulations (Liu et al., 2007; Zou, 2009).

Biological monitoring is a valid tool for assessing the internal exposure of a toxicant in the general population, and is particularly useful when applied in combination with other exposure assessment methods (Hertel et al., 2001). Thus, biological monitoring is conducted by collecting samples of human fluids and/or tissues (such as blood, urine, breast milk or hair) in order to detect exposure. There are different possible biological indicators for benzene exposure. Trans, trans muconic acid (t,t-MA), a urinary open-ringed metabolite constitutes a sensitive biomarker for benzene exposure, and can be used to differentiate populations exposed to external benzene levels of 0.5 ppm and smokers from non-smokers (Pezzagno et al., 1999).

The present work provides a quantitative assessment and validation of the individual exposure to benzene by using a new exposure modelling tool, the GPS based Exposure Model to Traffic-related Air Pollution (ExPOSITION). Also, a probabilistic approach based on the Johnson transformation system to characterize the variability of indoor concentrations in the predicted individual exposure is presented. The validation of the modelling approach is performed based on personal exposure measurements and biological monitoring data. For this purpose, exposure estimates obtained from personal monitoring and from biomarkers in urine samples collected during the daily activities of individuals were compared with exposure estimations in order to evaluate a feasibility of the proposed modelling approach.

6.2. Methodology

The Leiria urban area was selected in this study for the individual exposure modelling and monitoring. It is situated in the central part of Portugal and covering 8 sub-municipality units. The study domain (Figure 6.1) covering an area of 15 x 15 km^2 with 20m grid resolution for dispersion modelling and a complex terrain, containing about 34000 buildings considered as obstacles for the air dispersion modelling. The study period is focused from 21 to 25 of May 2012.

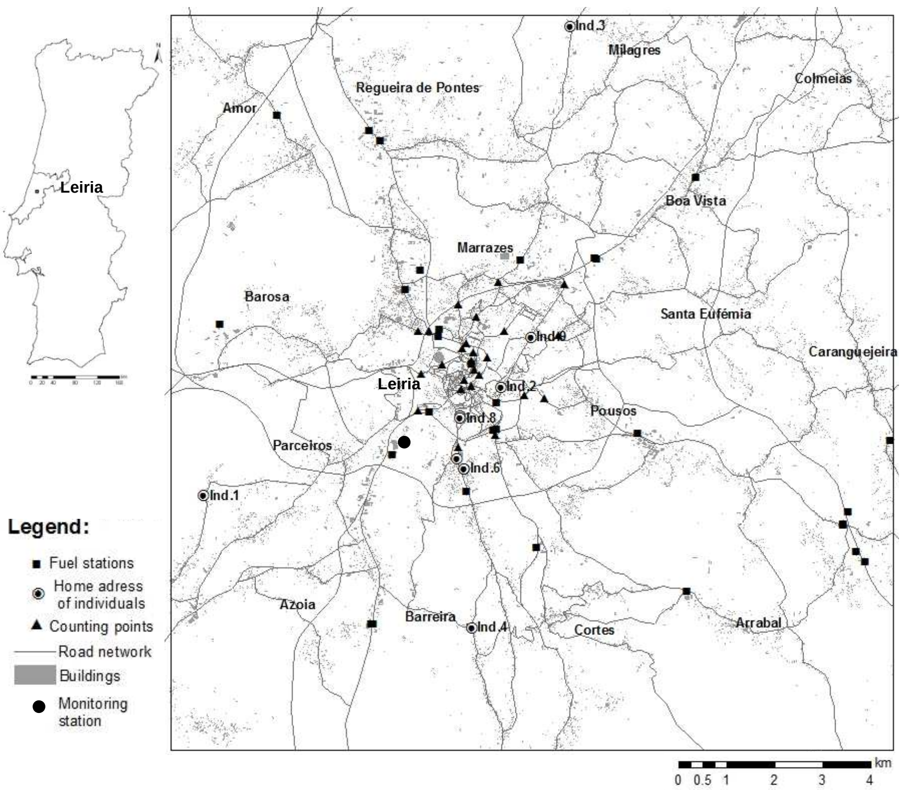


Figure 6.1. Study domain including road network, buildings, administrative units, and location of fuel stations, traffic counting points, air quality monitoring station and home adress of individuals.

The measurements campaign design and the human exposure modelling system applied in this study are described in the following sections.

6.2.1. Measurements campaign

The campaign has taken place in Leiria urban area, during 4 working days and 1 holiday. During this period the data from air quality monitoring station, individual exposure and biomonitoring, GPS trajectories and traffic counts were collected.

◢ Participating individuals

In this study, 10 healthy non-smoking adult volunteers were recruited to estimate individual exposure to benzene, providing their GPS trajectories by using mobile phones with GPS during their daily activities for the study period. Overall, the individuals selected in the framework of this study are office workers with only one exception that include a fuel station attendant. The selection of volunteers was performed without regard to age, sex, or ethnic background. Potential subjects were excluded if they were smokers, under 18 years old, unhealthy (e.g. had chronic respiratory or coronary disease or cancer), or their commute from home to work was not within the study area. Subjects resided in four different sub-municipality units of Leiria urban area: Parceiros, Barreira, Leiria and Milagres.

In order to validate the exposure model, personal and biological exposure to benzene was monitored during the usual daily activities of individuals and no restrictions on personal behaviour during the sampling time were imposed. However, due to the limited number of actively pumped personal samplers available, only 5 individuals were monitored during the same sampling time in order to validate the new exposure modelling approach.

◢ GPS Trajectories of individuals

The trajectories of the individuals were collected by TTGPSLogger tracking system (TTGPSLogger, 2012) installed on mobile phone providing second-by-second GPS data on the location of the volunteers. TTGPSLogger is a GPS logger software for Symbian S60 allowing to store detailed time-location information on geographic coordinates, speed and time during its use over the daily activities of individuals. This information was stored in a GPX file format that is compatible with Geographical Information System (GIS) presenting very useful to analyse the spatial distribution of large amount of GPS raw data collected.

◢ Traffic counts

For the traffic volume characterization, 25 counting stations spatially dispersed within the study area (Figure 6.1) were used providing information on traffic flow and distinguish between three vehicles categories (light vehicles, duty vehicles and motorcycles). Thus, a sample of data from the counting points were collected with intervals of 10 minutes during 1 hour and 30 minutes at morning peak hour, respectively, were considered in order to calibrate the transportation model.

◢ Air quality and meteorological parameters

For characterizing the ambient conditions, concentrations of several air pollutants including different fractions of particulate matter, ozone, sulphur dioxide, oxides of nitrogen, carbon monoxide, hydrocarbons, and meteorological parameters such as temperature, wind direction and wind speed were measured at one monitoring station located in sub-urban area of the city (Figure 6.1). Taking into account the objectives of the current study, only benzene concentrations obtained with Environment VOC71M (PID) analyser with a temporal resolution of 15 minutes are presented and analysed.

◢ Personal exposure monitoring

Simultaneously with individual's trajectories collection, the participants were carrying at their breathing zone actively pumped personal samplers to collect benzene concentrations during 24 hours, replacing the personal sampler through the day. Typically, for each day of the measurements campaign the personal samplers were substituted at 8 a.m., 2 p.m. and 8 p.m., obtaining a total of 37 personal samplers collected.

Benzene, toluene and xylene in air were analysed with an internal method based on ECA (1997). Briefly, air was collected on TENAX GR tubes using a personal air sampling pump (SKC Pocket pump) at a flow rate of 0.05 $l.min^{-1}$ for a period of approximately 8h (480 minutes). Analysis of compounds was performed by automatic thermal desorption coupled with gas chromatography fitted with flame ionization detector and one apolar column. Total Volatile Organic Compounds (TVOC) were quantified using the toluene response factor as already reported in Madureira et al. (2011). During the analysis of TVOC, concentrations of benzene, toluene and xylene were also determined. However, only benzene concentrations are analysed in the framework of this study.

◢ **Biological exposure monitoring**

Biological monitoring was carried out in parallel to the personal exposure monitoring with the purpose of analysing t,t-MA as biological indicator for benzene exposure. Thus, at the same personal samplers were replaced, spot urine samples were collected from each volunteer for t,t-MA analysis. Urine samples were collected in 30 mL polypropylene cups and transferred to laboratory in portable coolers containing ice packs and stored in the freezer at -20ºC. A total of 37 urine samples were collected (3 per subject per day).

t,t-MA was determined by a method described by Roma-Torres et al. (2006). The limit of quantitation for t,t muconic acid in urine was 50 µg.mL^{-1}. Concentrations obtained were corrected with the corresponding creatinine value. Creatinine was determined using CREAJ Gen2 kit (PN 04810716190, Roche Diagnostics) on COBAS INTEGRA 800 according to manufacturer instructions.

6.2.2. Human exposure modelling

The ExPOSITION model is developed to assess short (e.g. hourly, daily) and long-term (e.g. annual) inhalation exposures of the individuals to traffic-related air pollutants over urban spatial scale with high spatial-temporal resolution. For this purpose, air pollution concentrations (C_t) are estimated for different microenvironments j and combined with time t spent by individual i in each microenvironment using trajectories collected by mobile phones with GPS technology.

Personal exposure is characterised by ExPOSITION model in terms of time-weighted average exposure concentration calculated as following:

$$\overline{E}_i = \frac{1}{t_2 - t_1}\int_{t_1}^{t_2} C(x,y,z,t)_i \, dt \tag{6.1}$$

where $\overline{E}_i$ (µg.m^{-3}) is the average exposure concentration for person i, $C(x,y,z,t)_i$ (µg.m^{-3}) is the air pollutant concentration occurring at a particular point where the person i is located during the time t and spatial coordinate (x,y,z) and t_1 and t_2 (h) are the starting and ending times of the exposure event.

◢ Microenvironmental concentrations

Outdoor and several relevant indoor microenvironments are distinguished in the exposure model as presented in Table S.1 (see supplementary material, Section 6.5). Outdoor concentrations are estimated externally using atmospheric dispersion modelling as described in Section 6.2.3. For indoors and in-vehicle microenvironments it is assumed that: 1) within a microenvironment the pollutants are homogeneously distributed; 2) pollution levels in each microenvironment are related with outdoor pollution levels that occurring in the immediate vicinity to the microenvironment; 3) infiltration of outdoor pollution and contribution of indoor pollution sources is different for each type of microenvironments. In a general form the concentration $C(x,y,z,t)$ (µg.m^{-3}) for microenvironment j with a spatial coordinate (x,y,z) at time t is calculated taking into account the outdoor concentration $C(x,y,z,t)_{ambient}$ (µg.m^{-3}) at a neighbourhood cell, the outdoor/indoor ratio α_j (dimensionless) and the factor β_j (µg.m^{-3}) to characterize the additional contribution of indoor pollution sources:

$$C(x,y,z,t) = \beta_j + \alpha_j \times C(x,y,z,t)_{ambient} \tag{6.2}$$

However, due to the absence of European studies providing information on direct contribution of the indoor sources to the benzene concentrations in different microenvironments, β_j is described using a probabilistic approach. Thus, the variability of benzene indoor concentrations is characterised using random numbers generated from cumulative distribution function identified for each type of microenvironments. For this purpose the data reported by the PEOPLE project (Ballesta et al., 2006) are used in combination with Johnson transformation (Johnson et al., 1994) to fit the experimental data.

The Johnson system is widely used in the case of modelling data with an unknown distribution (Biller and Nelson, 2003) and has the flexibility to match any feasible set of values for the mean, variance, skewness, and kurtosis. This method is used in a wide range of applications, including human exposure studies (Flynn, 2006; 2007; 2010).

In this study, the Johnson system algorithm is implemented in MATLAB to generate a matrix of random numbers drawn from the distribution in the Johnson system that satisfies the four quantiles of the desired distribution. For this purpose, 1000 random numbers drawn from the appropriate distribution in the Johnson system were estimated to define β_j in the Equation (6.2) considering the percentiles of the experimental data reported for Lisbon (Ballesta et al., 2006), finding thus the values of the transformation coefficients that defines the corresponding distribution for each type of microenvironments (Table S.1, see supplementary material). The α parameter presented in the Table S.1 is estimated as a ratio between the median indoor concentrations to the median outdoor concentration reported for the measurements.

◢ Time-activity patterns

Location of the individuals in space and in time is required to estimate individual exposure in a combination with pollutants concentration fields provided by air pollution dispersion model. In this study, time-activity patterns were obtained from GPS trajectories.

The GPS dataset provide information on the locations in terms of coordinates (e.g. latitude and longitude) but contains no semantic meaning (Zhou et al., 2007) like the address or characteristics of location, i.e. type of microenvironments. Therefore, in order to obtain information on time-activity patterns the significant places and movement activities are extracted from the GPS raw data by ExPOSITION model using the trajectory data mining and analysed within GIS environment in order to overlay this information with other geo-spatial information.

For this purpose, several levels of GPS data processing are required in order to identify important patterns. Under this context, a preliminary processing of GPS data is implemented as a first step to “clean” the data by using an error-checking algorithm to remove invalid points, considering a measurement as valid if the GPS receiver is able to see at least four satellites and if the horizontal dilution of precision (HDOP) value is below 6. Also, incorrect entries of the travel speed are evaluated.

At next, the places where the individual was stopped for a certain time period are distinguished from moving activities, like driving a vehicle. This algorithm is iterative and it is based on searching for locations where the user has spent a longer time period depending thus on two scale parameters: a distance threshold and a time threshold (Li et al., 2008). Finally, it is necessary to discover which of these points belong to the same activity/place (significant places). For this purpose, a second level analysis based on a density-based clustering algorithm was implemented to group the points belonging to the same premises and to identify personally significant places.

In this study, “significant places” are considered as those locations that play significant role in the activities of a person, carrying a particular semantic meaning such as the living and working places, the restaurant and shopping mall, etc. Additionally, a “movement activity” is distinguished taking into account location change over time which can be aggregated by the purpose of the trip of an individual.

These data are further analysed within a GIS for classification of microenvironments and to obtain information on time-activity patterns. For this purpose, a geoprocessing of GPS data is performed using ModelBuilder module provided by ArcGis10 (Allen, 2011). The geoprocessing of GPS data is accomplished by considering analytical functions and several predefined criteria based on speed, time and spatial location register for the trajectory points to classify the significant places and movement activities to three activity categories: indoor, outdoor and in vehicle travel. The detailed GIS-maps are used to identify and to classify the microenvironments.

This detailed time-activity patterns for each individual will be linked with the pollutants concentration fields varying in space and in time provided by air pollution dispersion model described in the next section, producing exposure estimates within distinct microenvironments.

6.2.3. Transport, Emission and Air quality modelling

Air quality modelling allows establishing the relationships between current emissions and current air quality at particular locations. Information on variability of air pollutant concentrations is essential for the exposure quantification and these data may be provided by any modelling tools if it is compatible with ExPOSITION requirements in terms of spatial and temporal data resolution.

In the present study, road traffic and vehicle refuelling at fuel stations were considered as the main outdoor local emission sources of benzene. In this perspective, the characterization of hourly emissions from road traffic sources and vehicle refuelling required by the air quality model was performed.

In order to quantify transport activity data required by the road traffic emissions model, the classic, four-step model was used (Ortúzar and Willumsen, 2006). This model consists of four sequential submodels: trip generation; trip distribution; modal split; and traffic assignment. It determines the total trips generated (produced and attracted) in each one of the 104 zones into which the study domain was divided, distributes them to the other zones (104 x104 origin destination-pairs), allocates them to the different transport modes available, and finally assigns the vehicles to the road network. Trip generation and distribution was made based on the results of a previous study (Pinto et al., 2008), updated with recent socio-economic data and the traffic data obtained for the 25 counting stations (in Section 6.2.1.). Car traffic assignment was carried out according with the Wardrop principle – at equilibrium drivers cannot improve their travel times by changing routes

(Sheffi, 1992). Calculations were made with the TransPlan software (Santos and Antunes, 2005).

Hourly traffic emissions were estimated by Transport Emission Model for Line Sources (TREM). The emission factors considered by TREM depend on average speed, fuel type, engine capacity and emission reduction technology. A new version TREM-HAP (Transport Emission Model for Hazardous Air Pollutants) prepared to calculate HAPs emissions (Tchepel et al., 2012) has been used to provide inputs for AUSTAL2000 dispersion model.

The vehicle refuelling emissions considered in this study were quantified based on the CONCAWE methodology (CONCAWE, 2009). Vehicle refuelling emissions come from vapours displaced from the automobile tank by dispensed gasoline and from spillage. Thus, the emission of the pollutant p (E_p (kg)) for each fuel station i is estimated as following:

$$E_p = e_{pi} \times V_i \times TVP \qquad (6.4)$$

where e_{ip} is the emission factor (kg. m^{-3}.kPa^{-1}) for pollutant p and fuel station i; V_i is the volume of gasoline dispensed (m^3) for each fuel station i and TVP is the True Vapour Pressure of gasoline at storage temperature (kPa) (CONCAWE, 2009).

In order to calculate the atmospheric dispersion of benzene, the AUSTAL2000 dispersion model was applied in the current study allowing to establish relationship between emissions and air quality, and to provide hourly pollutants concentration fields. AUSTAL2000 is the official reference air dispersion model of the German Regulation on Air Quality Control for short-range applications and it is based on Lagrangian approach that simulates the dispersion of air pollutants by utilizing a random walk process (Janicke and Janicke, 2002; Janicke, 2004). The model system includes a diagnostic wind field model to account for terrain profile and/or buildings structures. Additionally to the detailed input data on emissions, a continuous time series of meteorological parameters, including wind direction, wind speed and atmospheric stability are required by AUSTAL2000.

To characterize air pollution related with non-traffic sources and/or transported from outside of the modelling domain, the background pollution levels were characterised. For this purpose, observations from the fixed monitoring station were used and processed to remove the local noise from the air quality time series in accordance with Tchepel and Borrego (2010) and Tchepel et al. (2010).

6.3. Results and Discussion

In this section, the results obtained from the measurements campaign and from modelling are presented and discussed.

6.3.1. Transportation and Emissions data

In order to estimate human exposure to benzene, hourly emissions from refuelling in fuel stations and from road traffic sources were estimated and inputted into the AUSTAL2000.

The spatial variations in traffic flows obtained from the transportation model and considered in hourly traffic emissions estimation is presented in Figure 6.2a evidencing higher traffic flow values for main urban area entrances roads.

Figure 6.2b illustrates the spatial variations in hourly traffic-related emissions and hourly automobile refuelling emissions across the study area obtained by linking emissions outputs to GIS maps. As could be seen in the figure, the largest contribution of benzene to the ambient air levels locally is the road traffic source contribution, evidencing as expected a spatial distribution of emissions similar to traffic flow observed for the study domain (Figure 6.2a).

a) b)

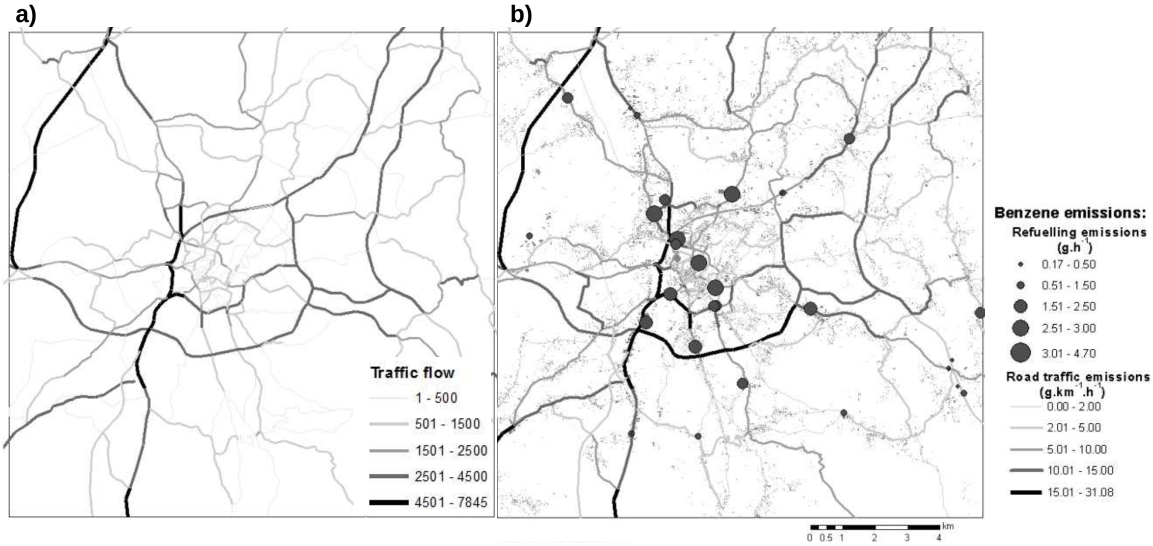


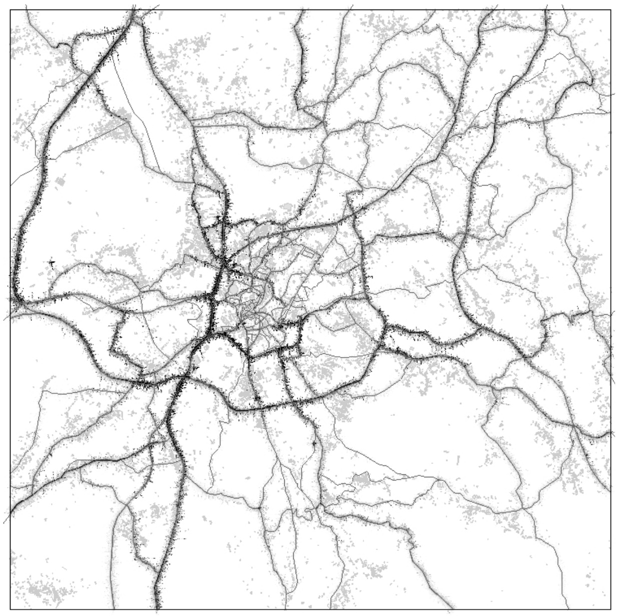
Figure 6.2. Spatial distribution of a) traffic flow at the morning peak hour and; b) hourly benzene emissions from fuel stations and road traffic sources.

6.3.2. Air quality, meteorological data and time-activity patterns

As mentioned in Section 6.2.1 the outdoor benzene concentrations and meteorological data were monitored in a sub-urban location at one fixed monitoring station (Figure S.1, see supplementary material).

The spatial distribution of the air pollutants concentration obtained by dispersion modelling AUSTAL2000 is presented in Figure 6.3a showing non-homogeneous distribution of benzene levels within the study domain.

a)



b)

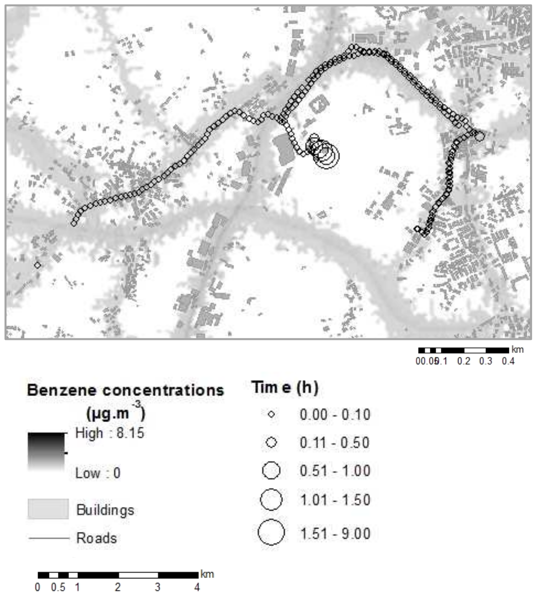


Figure 6.3. a) Spatial distribution of daily benzene concentrations related with emissions from modeled sources in the study domain; b) An example of time spent by the individual in each microenvironment during a typical working day.

The analysis of the results indicates that although emissions from road traffic will determine the overall pattern of benzene concentrations related with distribution of main network, important hot-spots of high concentration are also located in close proximity to gasoline fuel stations. Also, time-activity patterns obtained for one of the individuals are presented in the Figure 6.3b as an example, evidencing the variation of benzene concentrations in space and in time provided by air pollution dispersion model and the influence of time spent in each microenvironment type.

6.3.3. Individual exposure modelling

The individual exposure assessment performed by the ExPOSITION model is presented in this section. In order to better understand the contribution of different microenvironments to the individual exposure to benzene obtained during the study period, several statistical parameters calculated based on data for 10 individuals, including average individual exposure, 5th and 95th percentile and extreme values obtained from the ExPOSITION model were analysed (Figure 6.4a).

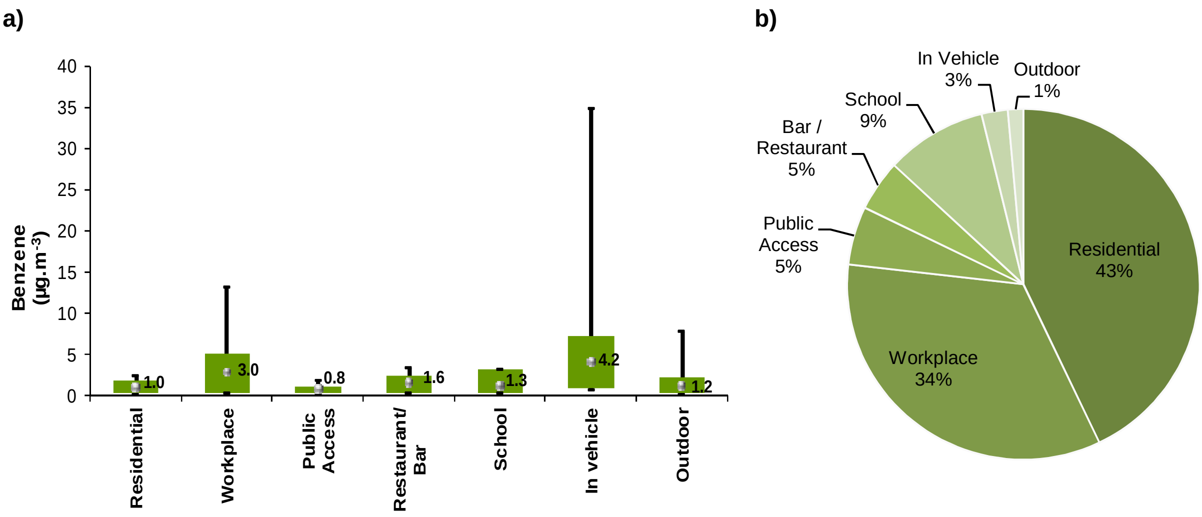


Figure 6.4. a) Exposure concentrations for benzene (µg.m^{-3}) in different microenvironments; b) Time-distribution of time-activity patterns of all individuals.

A considerable variability in the benzene exposure concentration in each microenvironment type is evidenced in Figure 6.4a, namely for “in vehicle” and “workplace” microenvironments, showing thus the importance to consider the distinct microenvironmental concentrations in individual exposure modelling. The higher exposure concentrations estimated for workplaces (about 44% to the total daily values) evidence the important contribution of their indoor sources to personal exposure. This fact is also related with the proximity of the working places (offices) of the considered individuals to urban roads with intensive traffic, as well as contribution of benzene pollution levels obtained for the fuel station attendant. On the other hand, exposure concentration calculated for residence are characterised by smaller variability range. However, exposure levels at residences represent a great relevance due to the time spent (about 43%) by the individuals during their daily activities (Figure 6.4b).

In order to better analyse the individual exposure obtained during the study period (5 days), the temporal variation of the exposure concentration modelled for the 10 individuals, whose GPS trajectories were collected, was analysed as presented in Figure S.2 provided in supplementary material. Also, the temporal variation of the outdoor background concentrations obtained from the fixed monitoring station during the study period is presented (Figure S.2).

6.3.4. Validation of the individual exposure model

In order to evaluate a feasibility of the proposed modelling approach, the ExPOSITION model predictions and exposure measurements obtained from personal monitoring and from urinary biomarker in urine samples collected during the daily activities of the 5 individuals are presented and analysed in this section.

For evaluation of the modelling approach by means of comparison with direct measurement obtained from personal monitoring, different statistical indicators were estimated (Figure 6.5a). The analysis is presented considering the values obtained from several personal samples (37 values) collected during the daily activities of individuals and model outputs averaged over the same sampling period. As could be seen from Figure 6.5a, a good agreement between the personal exposures predicted by the model and the data from actively pumped personal samplers is obtained. The ability of the model into follow the temporal variability of personal exposure measurements collected during daily activities of the different individuals is evidenced in Figure 6.5a, reflecting thus the temporal variability impact of meteorological conditions and emissions data in the predicted individual exposure. The Pearson's correlation coefficient of 0.66 with a P-value < 0.0001 and 95% confidence interval of 0.42 to 0.82 between two dataset confirms the model capability to describe the exposure variations in time, in space and between the individuals. In addition, the positive fractional BIAS value of the 0.32 shows that the exposures are over-estimated by the proposed model, being within the range of the acceptable values ((-2) to 2) and very close to the ideal value of 0. The good performance of the exposure model is also evidenced by the low value of the normalized mean squared error (0.8), as well as 71% concentrations are predicted within a factor of two.

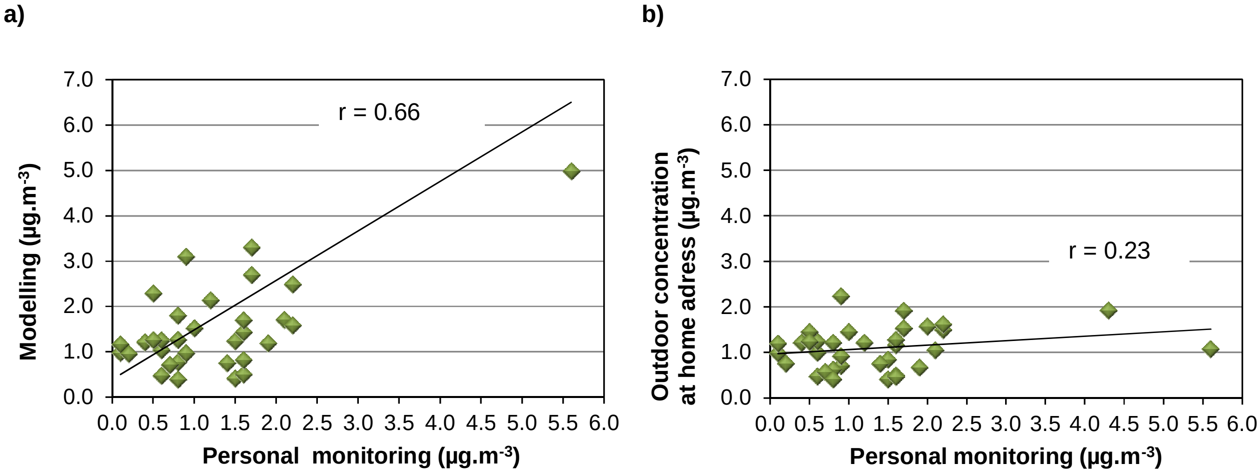


Figure 6.5. Scatter plot of benzene individual exposure obtained by: a) the modeling approach and by personal monitoring (µg.m^{-3}); b) the modeling approach based on the home address and by personal monitoring (µg.m^{-3}).

In several studies, personal exposure based on home address is considered as a good exposure indicator. Therefore, the feasibility of this exposure metric is presented and analysed in this study based on personal exposure measurements collected during daily activities (Figure 6.5b). Thus, as evidenced in the figure and confirmed by the Pearson's correlation coefficient of 0.23 (P=0.1670, 95% CI -0.10 to 0.53) between two dataset, there is a poorer agreement between the personal exposures estimate at residence place and the data obtained from personal sampler, which suggests that the proposed modelling tool based on the trajectory analysis presents as a more consistent approach to address the temporal and spatial variability of the personal exposure in urban areas.

For evaluation of the modelling approach, several statistical parameters, including daily average exposure concentration, 5th and 95th percentile are also analysed for each individual and compared with direct measurements obtained from actively pumped personal samplers as presented in Figure 6.6.

Overall, as could be seen in Figure 6.6, the range of personal exposure levels obtained from the ExPOSITION model is in agreement with the exposure measurements showing only exception for the fuel attendant (individual 4) that evidencing an overestimation of the exposure levels by the model for this individual. Model overestimation of evaporative benzene emissions attributed to refuelling is one of the plausible causes for the exposure overestimation for the individual 4 during the working hours.

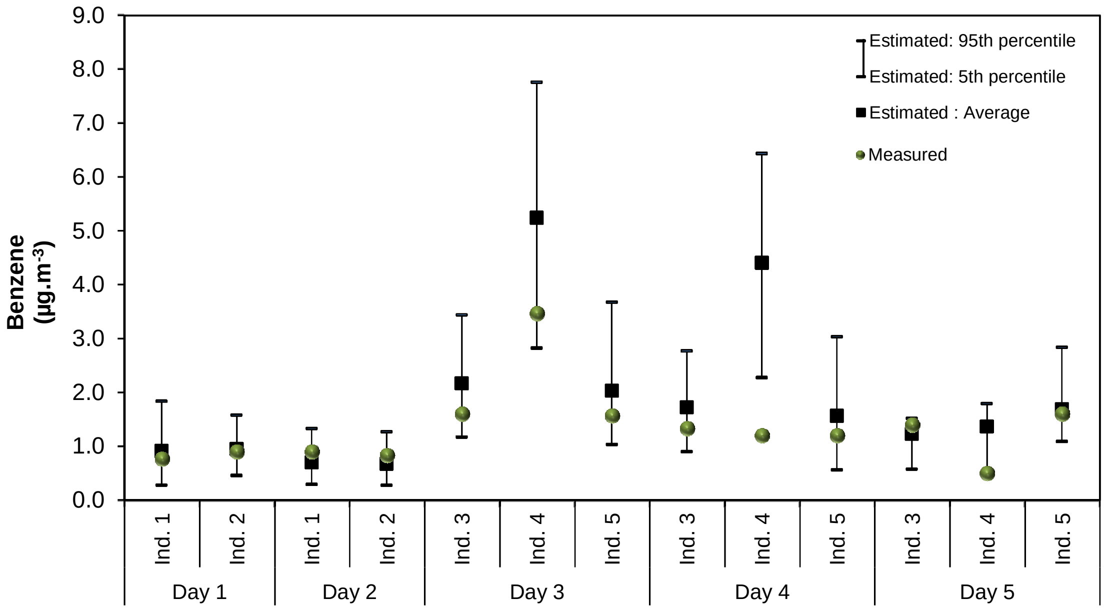


Figure 6.6. Relation between daily average exposures to benzene provided by the model and measurements of individual exposures obtained by personal monitoring.

Individual exposure estimated by the model and measured from personal monitoring are also compared with biomonitoring data using trans,trans-muconic acid (tt-MA) in urine as benzene biomarker (Figure 6.7). In this analysis, daily average values were used in order to cover the temporal representativeness of biomonitoring samples.

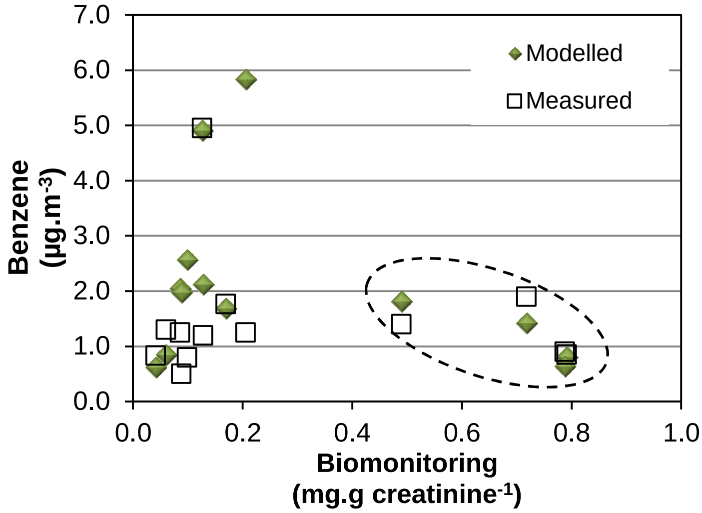


Figure 6.7. Scatter plot of benzene individual exposures measured and provided by the model ($\mu g.m^{-3}$) and concentrations of tt-MA in urinary samples (mg.g creatinine^{-1}).

As could be seen from the Figure 6.7, a cluster of points with notably different behaviour was identified in the biomonitoring data presenting no direct correlation with both the exposure model and personal monitoring. This fact could be related with external factors provided by other exposure routes (e.g. dietary), not just air, that are only reflected by the biomonitoring data. Thus, excluding the 4 points with no direct correlation with personal measurements, a Pearson's correlation coefficient of 0.74 (P=0.0238, 95% CI 0.14 to 0.94) between the daily average estimated values and measured from biomonitoring is obtained, indicating thus a good agreement between two dataset.

As mentioned previously, biomarkers estimates consider all exposure routes and sources over time. Although this is a main advantage in many situations it can also make difficult data interpretation. Noteworthy, out of the amount of absorbed benzene by humans, it has been estimated that only approximately 2% is eliminated as t,t-MA (Senzolo, 2001). Moreover, in addition to benzene exposure, smoking, genetic susceptibility, coexposure to toluene and pregnancy, intake of the preserving agent sorbic acid, which is a widely used preservative in food products, can influence the levels of urinary t,t-MA (Scherer, 1998). In this study, we can exclude the first factors but because no information was collected on food and drink intake in the study subjects, the contribution of sorbic acid and its salts in the excretion of t,t-MA could not be properly evaluated. Furthermore, the higher levels of t,t-MA were obtained in samples collected after mealtime. Previous studies have shown (Pezzagno et al., 1999) that after oral administration of sorbic acid contained in food may account for urinary t,t-MA levels similar to those found due to occupational exposure to benzene.

6.4. Conclusions

In this study, a comprehensive approach to quantify individual exposure to benzene in urban areas with high temporal and spatial resolution is implemented based on a new exposure modelling tool ExPOSITION. An application example and validation of the modelling approach against personal exposure measurements and biological monitoring data is presented and discussed. Overall, the daily average exposure to benzene predicted by the ExPOSITION model correspond to 1.6 $\mu g.m^{-3}$ in terms of the mean value for all individuals and 0.8 to 2.7 $\mu g.m^{-3}$ in terms of 5^{th} to 95^{th} percentiles. Individual exposure is particularly sensitive to high spatial and temporal variations of the pollution levels, emphasizing the importance of the indoor microenvironments and hot spots contribution that suggest limited representativeness of background concentrations obtained from point measurements.

The evaluation of proposed modelling approach by means of comparison with direct measurements collected for the selected individuals performed in this study indicates that there is a good agreement of the model results with personal monitoring and t,t-MA considered as biological indicator of benzene exposure showing a Pearson's correlation coefficient of 0.66 ($P<0.0001$, 95% CI 0.42 to 0.82) and 0.74 ($P=0.0238$, 95% CI 0.14 to 0.94), respectively.

The modelling approach presented in this work provides more consistent results in comparison with the personal exposure estimates based on home address outdoor concentrations, as demonstrated by the lower Pearson's correlation coefficient of 0.23 between personal exposure based on home address and the data from actively pumped personal samplers. Thus, the proposed modelling tool based on the trajectory analysis presents as a more consistent approach to a better understanding of exposure by establishing source-receptor relationship and by explicitly addressing the temporal and spatial variability in the exposure.

6.5. Appendix. Supplementary data

Table S.1.

Table S.1. I/O ratio and coefficients used to define the Johnson distribution for different microenvironments.

Microenvironment	α	Type of distribution*	Coefficients			
			γ	δ	ξ	λ
Residence	0.92	SU	-0.6649	1.1791	-0.1808	0.4686
Vehicle	2.42	SB	1.1976	0.4857	-0.0845	1.7120
Office	1.55	SU	-2.2909	2.3231	-0.6518	0.6161
School	1.11	SB	0.5003	0.1903	-0.9400	2.6853
Public access	0.42	SU	-0.5429	0.5088	-0.0559	0.1075
Restaurant/Bar	1.16	SB	0.4686	0.6534	-0.6524	2.8782

*SB - Logistic transformation (bounded); SU - Hyperbolic sine transformation (unbounded); γ and δ - shape parameters; ξ - location parameter; λ - scale parameter

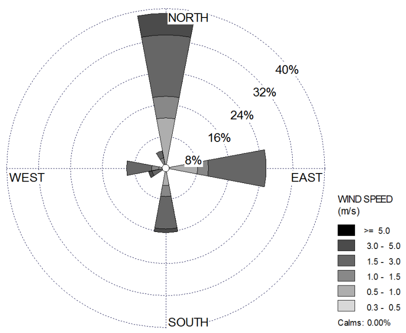
The Johnson transformation system can be written as:

$$X = \gamma + \delta \cdot \Gamma\left(\frac{(Z-\xi)}{\lambda}\right)$$

where X is the random variable X whose distribution is unknown; Z a standard normal random variable with mean 0 and variance 1 so that $Z \sim$ N(0, 1); y and δ are shape parameters, λ is a scale parameter, ξ is a location parameter, and Γ is the transformation whose form defines the four possible distribution families in the Johnson translation system known as identity (SN), exponential (SL), logistic (SB), and hyperbolic sine transformations (SU).

Figure S.1.

a)



b)

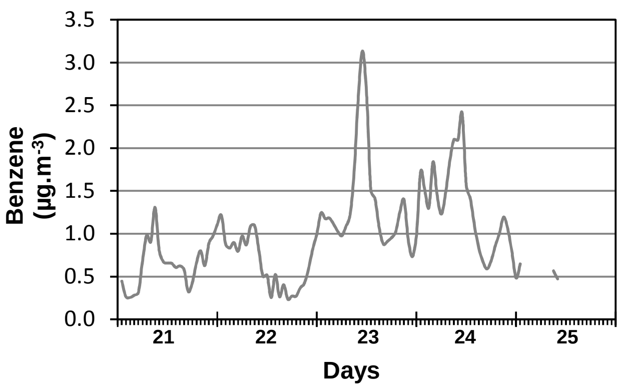


Figure S.1. a) Hourly wind speed obtained from measurements as a function of wind direction; b) temporal variation of hourly average background benzene concentrations.

The outdoor benzene concentrations and meteorological data monitored in a sub-urban location at one fixed monitoring station during the sampling period are presented in Figure S.1. As could be seen from the Figure S.1a, the higher wind intensities are achieved with winds blowing from the North, which is also the predominant wind direction, although there is a significant contribution of the East direction. As regards the variation throughout the day, generally the wind speed gradually increases, reaching maximum values between 2 p.m. and 5 p.m. and minimum values during the night.

The time series of hourly background concentrations of benzene measured from fixed monitoring station is presented in the Figure S.1b, evidencing a pronounced diurnal variation for benzene concentrations during the study period. The lower concentrations of benzene are observed during the holiday, day 22, mainly for the time period between 12 p.m. to 6 p.m.. Additionally to the lower emissions, high values of wind speed were observed during the same time period that influences the dispersion conditions and consequently benzene concentrations. On the other hand, the highest values are reached during the 3rd day of the campaign period, achieving hourly maximum value of 3.10 µg.m^{-3} and 1.34 µg.m^{-3} of daily average concentrations.

Figure S.2.

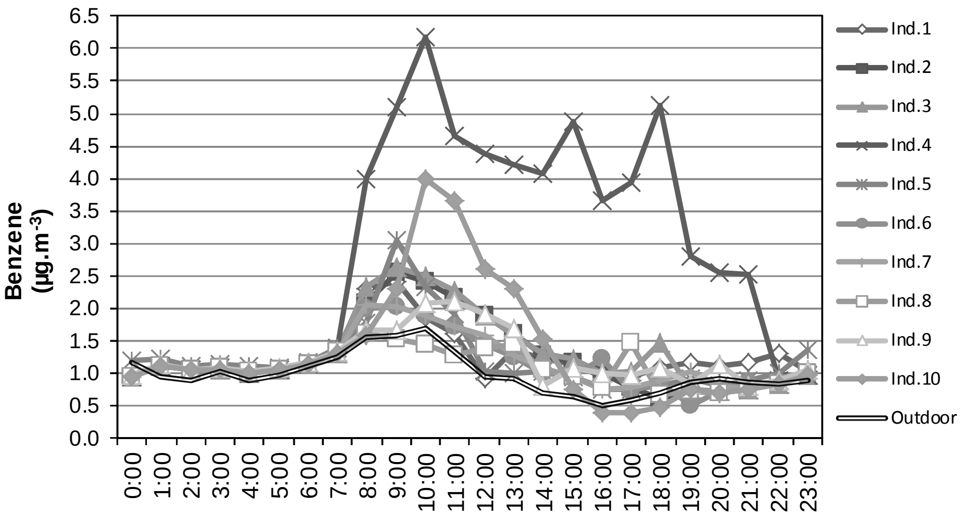


Figure S.2. Temporal variation of individual exposure estimates and measurements of outdoor background concentrations for benzene (µg.m^{-3}).

The temporal variation of the exposure concentration modelled for the 10 individuals and the outdoor background concentrations obtained from the fixed monitoring station during the study period is presented in Figure S.2. The results suggest that the 10 individuals are exposed to different benzene concentrations during their daily activities, and a significant variability in benzene exposures across the individuals is evident. Moreover, it is clear in Figure S.2 that the benzene background concentrations measured at monitoring station are significantly lower than the exposure concentrations estimated for the individuals. Therefore, point fixed background observations may not be representative to describe the range of exposure to benzene. The individual exposure concentrations during night time (until 7 a.m. approximately) when the people stay in a residence presents a similar trend with the outdoor background concentrations. However, throughout the day and depending on the daily activity of the individual the hourly average exposure concentrations tend to be greater in magnitude and more variable than background pollution levels.

As expected, the individual 4 (fuel station attendant) is affected by the highest exposure concentration values with a peak value at 10 a.m., 3 p.m. and 6 p.m. of about 6.2 $\mu g.m^{-3}$, 4.9 $\mu g.m^{-3}$ and 5.1 $\mu g.m^{-3}$, respectively. This is related with the highest concentrations and the time spent in the workplace during this time period.

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CHAPTER SEVEN

7. GENERAL CONCLUSIONS

The main purpose of the research presented in this dissertation was to develop a consistent approach and a new exposure modelling tool to estimate individual exposure to traffic-related hazardous air pollutants with high spatial and temporal resolution based on an innovative approach for trajectory analysis of the individuals. This research performed through a series of spatial analysis and modelling approaches intends to contribute to an improved knowledge regarding personal exposure to air pollution in the urban environment. The main achievements are presented and organized in seven chapters starting with the overall introduction to the particular topics, human exposure, urban air pollution, exposure-related health effects, human mobility patterns and technological resources, and their relationships.

7.1. Summary of Research and Findings

The evidences of health effects related to exposure to air pollution at levels usually experienced by individuals in urban areas were analysed and established. The modelling results suggest a significant potential health benefits by meeting the air quality limit values (2008/50/CE) for short-term PM10 exposure in one of the most affected areas by higher concentrations in future climate, Porto Metropolitan Area. The study pointed to the potential annual reduction of 3.2 (95% CI 2.24 – 4.18) deaths.100 000 inhabitants^{-1} due to cardiovascular diseases and 2.12 (95% CI 0.53 – 3.95) deaths.100 000 inhabitants^{-1} due to respiratory diseases, by meeting the air quality limit values (2008/50/CE) for cumulative short-term (40 days) exposure to PM10. Moreover, an improved methodology to process population statistics taking into account daily average population mobility and filtering of air quality time series to improve representativeness of measurements was implemented. The results suggest that the potential health benefits related with the reduction of air pollution levels for the study population estimated by this novel approach are 50 – 56% higher than those provided by the traditional approach (exposure estimate without human mobility). These findings suggest that human mobility and inhomogeneity of air pollution levels determine human exposure to urban air pollutants, and should be considered to characterize human exposure for an improved health impact assessment. Moreover, the distinct results obtained with and without population mobility are strongly influenced by the input data on population mobility and air pollution spatial variation considered in the analysis thus showing the sensitivity of the short-term risk assessment methodology to these parameters.

Also, health risk within urban areas was evidenced under climate-induced changes in air pollution levels. The results obtained in this study revealed that climate change alone will deeply impact the PM10 levels in the atmosphere, affecting consequently all the Portuguese districts with pronounced negative effects on human health, mainly in major urban areas, such as Porto and Lisbon. The short-term variations in the PM10 concentration under future climate will potentially lead to an increase of 203 premature deaths per year in Portugal, achieving the most significant increase in premature deaths in Porto area, corresponding to approximately 8%. Also, the study pointed to 81% of cases attributed to future pollution episodes with daily average PM10 concentration above the current legislated value (50 $\mu g.m^{-3}$). In addition to importance of indirect effects of climate change on human health, this study also highlights the significant contribution of pollution peaks in urban areas to acute exposure, despite their low frequency. Given the little information concerning the impact of environmental factors on human health that has been published for Portugal, these outcomes provide important information to support local and national policy related with air pollution and human health issues.

For a comprehensive understanding of exposure to traffic-related air pollution in urban areas and consequent health effects, the quantification and characterization of traffic-related emissions with high spatial and temporal resolution was performed by developing a modelling approach for quantification of hazardous air pollutants emissions related to the traffic activity in urban areas. The results obtained by application of the Transport Emission Model for hazardous air pollutants (TREM-HAP) pointed different trend taking into account the seasonal variations (summer and winter periods) on total daily emissions of traffic-related hazardous air pollutants for the analysed urban area. Benzene emissions are 17% higher at winter time due to important contribution of cold starts while other traffic-related air pollutants are mainly affected by seasonal changes in the traffic volume observed for the study area, resulting in higher emissions during the summer period. Also, a probabilistic emission inventory for traffic-related air pollutants considering different road types was obtained for an urban area. Several statistical parameters were analysed for the selected pollutants, evidencing that PM2.5 and benzene have the largest uncertainty in the absolute daily emissions. In addition, highly uncertain emission data were obtained for the urban roads. Oppositely, emissions calculated for highways were generally characterised by a very small uncertainty (less than ±5%) except for PM2.5 (–16% to +9%). This information on spatial distribution of the traffic-related air pollutants emissions for each road segment are essential for air quality modelling and further exposure assessment studies. Also, this tool opens a possibly to analyse human exposure to traffic-related air pollution in urban areas using probabilistic approach, integrating transportation policy definition with the air quality assessment and human exposure assessment.

Through this work, the spatial and temporal heterogeneity of air pollution levels that characterizes the urban environment was evidenced and identified as a major issue for study of exposure to urban air pollution. Thus, based on enhanced technological resources, namely GIS and GPS, a new modelling tool for quantification of short and long-term exposure to urban air pollution at the temporal and spatial scale required to estimate exposure at the individual level was developed for better understanding of exposure-related health effects to urban air pollution. The development of the GPS based Exposure Model to Traffic-related Air Pollution model (ExPOSITION) constitutes one of the major results of this work. The ExPOSITION model was developed based on a novel approach for trajectory analysis of the individuals collected via mobile phones with GPS technology and air pollution modelling with high spatial-temporal resolution within distinct microenvironments. Thus, one of the innovative aspects of this work was the development and implementation of an algorithm based on trajectory data mining analysis and geo-spatial analysis within GIS to process the GPS trajectories and extract the time-activity patterns of individuals, enabling to locate and classify microenvironments frequented by the individuals during their daily activities, as required for the exposure assessment. In addition, two different approaches were considered to characterize the pollution levels in these several microenvironments distinguished in the ExPOSITION model (i.e. residence, other indoors, outdoors, and in-vehicle). Thus, outdoor concentrations are estimated using atmospheric dispersion modelling and different modelling tools may be used to provide this external information for ExPOSITION. For indoors and in-vehicle microenvironments a probabilistic approach based on Johnson system of distributions was implemented as an integrated part of ExPOSITION algorithm to characterize the variability of indoor concentrations in the predicted individual exposure.

In order to characterize an individual’s contact with a given urban pollution levels at different microenvironments, the ExPOSITION model was applied to Leiria urban area to quantify the short-term individual exposure (1 day) to PM2.5. To achieve this purpose, hourly PM2.5 emissions from road traffic were estimated by TREM-HAP and PM2.5

concentrations hourly simulations were conducted with AUSTAL2000 model taking into account hourly meteorological conditions and background concentrations given by the nearest background air quality monitoring station. The results obtained by the time-activity pattern discovery sequence, based on trajectory data mining and geo-spatial analysis within GIS, highlights the added value of this innovative approach for exposure assessment. For instance, analysing the results achieved during the several levels of GPS data analyses in case of one of the individuals, 30179 collected GPS raw points resulted in 295 important locations that are linked with the pollutants concentration in distinct microenvironments to assess his individual exposure. Such results also indicate that this approach could overcome some limitations related with the analysis of GPS raw data and its implications for human exposure assessment, thus allowing to identify and classify time-activity patterns based on raw GPS tracking data at the spatial and temporal scale required for exposure assessment.

Additionally, the individual exposure estimates provided by the ExPOSITION model give relevant information regarding the importance of indoor microenvironments' contribution to the daily individual exposure to PM2.5 in urban areas, particularly the residence (51%), thus stressing that individual exposure depends not only on the pollution levels but also on the time spent in the microenvironment during the individual´s daily activities. In addition, it was possible to verify that the variability in the PM2.5 exposure concentration in each microenvironment type is significant showing the importance to consider this variability in individual exposure modelling. Overall, the daily average exposure to PM2.5 predicted by the ExPOSITION model correspond to 10.6 $\mu g.m^{-3}$ in terms of the mean value for all individuals and 6.0 – 16.4 $\mu g.m^{-3}$ in terms of 5^{th} – 95^{th} percentiles. Comparing the mean value obtained by the model and estimated from air quality measurements at a fixed point (11 $\mu g.m^{-3}$), an agreement between the approaches was evidenced. However, the ExPOSITION model reveals additional inter and intra-variability of individuals' exposure levels that is essential for health impact assessment and epidemiological studies, suggesting limited representativeness of air quality concentrations obtained from point measurements to characterize individual exposure to urban air pollution. In this context individual time-activities patterns and time spent at different microenvironments during the day should be of prime concern additionally to the variability in the urban pollution levels.

The validation of the proposed modelling approach was performed for individual exposure to benzene in the urban environment against personal and biological exposure measurements collected during a measurements campaign. In this study, a modelling cascade including transportation-emission-dispersion modelling was implemented to characterise the outdoor pollution within Leiria urban area. Overall, as identified for PM2.5

exposure, the exposure modelling results in the different microenvironments pointed that the personal exposures to benzene tend to be greater in magnitude and more variable than the corresponding ambient concentrations within urban areas. The average exposure estimated by the ExPOSITION model was 1.5 times higher than average ambient background concentration observed at point monitoring station during the campaign, emphasizing the importance of the indoor microenvironments and hot spots contribution to benzene exposure. The evaluation of proposed modelling approach by means of comparison with direct measurements indicates that there is a good agreement of the model results with personal monitoring and biological monitoring using t,t-MA as biological indicator of benzene exposure showing a Pearson's correlation coefficient of 0.66 ($P<0.0001$, 95% CI 0.42 to 0.82) and 0.74 ($P=0.0238$, 95% CI 0.14 to 0.94), respectively. Also, the results indicate that the ExPOSITION model validated in this study presents as a more consistent approach to a better understanding of exposure, providing more consistent results in comparison with the personal exposure estimates based on home address outdoor concentrations, as demonstrated by the lower Pearson's correlation coefficient of 0.23 between personal exposure based on home address and the data from actively pumped personal samplers.

The novel methodology proposed in this work and based on the system of integrated modelling tools (transportation, emission, air quality and exposure models) and advanced technological resources (GPS and GIS) allows to characterize the complexity in the spatial variation of exposures among the different individuals and delivers main statistics on individual's air pollution exposure. Moreover, this methodology contributes to exposure research by emphasizing individual time-activity patterns in the individual air pollution exposure context, providing thus new insights into individual exposure to urban air pollution and its effects on human health.

7.2. Future research

An improvement of the methodology for individual exposure quantification is a continuous task. Further developments of ExPOSITION model could include new health-relevant metrics of the particle mass. Smaller particles bear a larger toxic potential while contributing relatively little to the PM2.5 or PM10 mass. Therefore, future research could be focused on the individual exposure to PM1 and other nano-sized particles. Also, further efforts should be made to characterise the components of particulate matter (PM), such as trace elements for individual exposure assessment.

Evaluation of time-activity patterns discovery sequence from raw GPS data is a complex task. More developments are required mainly to infer about the travel mode used by the individuals from their GPS trajectory collected. Future developments should be focus on more refined classification based on a combination of GPS records of these travel modes and related supplemental GIS information (e.g. bus and train routes) to be used for individual exposure assessment.

For future research, the presented approach could be extended to a near real-time information system for individuals by a web-based implementation of the model. Based on their uploaded time-activity patterns, a user without expert knowledge in exposure modelling could assess their own individual exposure, also identifying mitigation measures to regulate ambient concentrations specifically defined for this individual based on their personal behaviour and spatial distribution of the air pollution. This might also help to identify the "low-exposure" route, transportation mode and time for their journeys through a city.