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DOCTORAATSPROEFSCHRIFT

Air pollution exposure assessment through personal monitoring and activity-based modeling

Proefschrift voorgelegd tot het behalen van de graad van doctor in de verkeerskunde te verdedigen door:

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SUMMARY

Background and aims

Evidence is growing that a myriad of health effects, ranging from respiratory to cardiovascular and neurological effects, are causally linked to exposure to air pollution. Annually an estimated number of 2 million premature deaths can be attributed to air pollution worldwide. In recent years, society has become increasingly sensitive to individual risks, and thus data on the exposure to air pollution needs to be personalized. Historically exposure of the population has been calculated by simply multiplying pollutant concentrations with population density at the same location, ignoring population movement. Obtaining personal exposure measurements has been hampered by the cost and complexity of the analyzing equipment. But accurate exposure assessment is critical to further reduce exposure misclassification in epidemiological studies. Alternatively, the risk of erroneously attributing health effects to a statistically associated but toxicologically harmless pollutant will be diminished if exposure is mapped more truthfully.

The goals of this PhD project are firstly to demonstrate the feasibility of personal monitoring using innovative air quality monitors. Secondly, it will be shown that activity-based models are a promising new tool to model exposure to air pollution in populations; an existing framework was fundamentally adapted so that it can predict personal exposure.

The newly developed methodologies are demonstrated by applying them for the air pollutant black carbon (BC), that is identified as an important proxy for traffic-related pollution and is a component of particulate matter. Over the last decades BC concentrations have declined in developed countries, although the air has still moderate to heavy BC pollution. It has been shown that BC has important health effects in humans, and it is a potent climate warmer, resulting in its inclusion in several recent high level policy documents.

Materials and methods

Personal exposure to air pollution can be determined using indirect exposure models or through a direct approach (i.e. air quality measurements). The newly developed Flemish activity-based model FEATHERS is used to model the

activities that people perform (e.g. work, shopping, sleeping, traveling) as well as the times and locations. Land use regression (LUR) models are built using measurements on 63 fixed locations in Flanders to estimate air pollution levels with a high spatial and temporal resolution. In a LUR model statistical associations are developed between potential predictor variables and measured pollutant concentrations as a basis for predicting concentrations at unsampled sites. LUR predictions are then combined with a fixed indoor/outdoor factor for exposure in indoor environments. To estimate exposure in transport, a separate model is developed taking into account transport mode, timing of the trip and degree of urbanization. The modeling framework is validated with personal measurements. This monitoring campaign in 62 volunteers included the weeklong measurement of BC using micro-aethalometers, and the collection of diaries and GPS logs on an electronic platform. This resulted in more than 10,000 hours of data, and the registration of more than 1500 single trips.

Results and conclusions

Personal monitoring revealed that exposure to BC can differ between partners, living at the same address, by up to 30%. On average, homemakers are exposed to lower concentrations compared to full-time workers, mainly because of the difference in travel time and the elevated concentrations while traveling. In the study cohort, 6% of the time is spent in transport, but this accounts for 21% of personal exposure to BC and approximately 30% of inhaled dose. Unfortunately travel time proved to be an unsatisfactory predictor of personal exposure because of the many factors influencing concentrations in transport. Concentrations are highest in motorized modes (car, bus, light rail / metro), and lowest for active modes and trains; but exposure in active modes becomes more important when inhalation is taken into account. In-vehicle BC concentrations are elevated on highways and on urban roads, during rush hour and on weekdays, at speeds <30 km/h and >80 km/h, and increases with increasing traffic intensities on the roads travelled. The latter proved to be the major explanatory variable for in-vehicle BC concentrations, together with timing of the trip and urbanization. For cyclists and pedestrians exposure is influenced by time of day and degree of urbanization.

Personal exposure was modeled by using the FEATHERS model, hourly LUR models, an indoor air model, and an in-traffic exposure model; combined into the AB²C framework (Activity-Based modeling framework for BC exposure assessment). Building independent hourly LUR models resulted in R^2 values mostly smaller than the R^2 of the annual model (R^2=0.77), ranging from 0.07 to 0.8. Between 6 a.m. and 10 p.m. on weekdays the R^2 approximates the annual model R^2. Even though models of consecutive hours are developed independently, similar variables turn out to be significant. Using dynamic covariates instead of static covariates, i.e. hourly traffic intensities and hourly population densities from FEATHERS, did not significantly improve the models' performance.

The modeling framework AB²C is validated using time-activity diaries and BC exposure as revealed from the personal monitoring campaign with 62 participants. For each participant in the monitoring campaign, a synthetic population of 100 model-agents per day was generated with all agents having the same characteristics as each real-life agent. The AB²C model then calculates for each individual a distribution of potential exposures. Average personal exposure was estimated more accurately by AB²C compared to ambient concentrations as predicted for the home subzone; however the added value of a dynamic model lies at the moment primarily in the potential to detect short term and repeated peak exposures rather than modeling average exposures.

In summary, this dissertation demonstrates the potential, the advantages and the limitations of personalized exposure monitoring in a subset of the population. An activity-based exposure modeling framework was developed to estimate the exposure of individuals within a larger population. Several unique submodels were linked to result in the innovative exposure model AB²C.

SAMENVATTING

Achtergrond en doelstellingen

Er zijn steeds meer aanwijzingen voor een causale relatie tussen blootstelling aan luchtvervuiling en gezondheidseffecten bij de mens; zowel respiratoire aandoeningen als cardiovasculaire en neurologische effecten kunnen gelinkt worden aan luchtvervuiling. Wereldwijd kunnen jaarlijks naar schatting 2 miljoen vroegtijdige overlijdens toegeschreven worden aan een slechte luchtkwaliteit. Recent is de maatschappij meer gevoelig geworden aan individuele risico's, en dus moeten ook data over blootstelling aan luchtvervuiling gepersonaliseerd worden. In het verleden werd de blootstelling van een populatie bepaald door concentraties simpelweg te vermenigvuldigen met de bevolkingsdichtheid op die locatie; het feit dat mensen niet altijd op dezelfde locatie verblijven werd gemakshalve genegeerd. Het meten van persoonlijke blootstelling werd beperkt door de beschikbaarheid, de kost en de complexiteit van meetapparatuur. Een nauwkeurige inschatting van blootstelling is nochtans belangrijk om misclassificatie van blootstelling te verminderen in epidemiologische studies. Anderzijds vermijden we zo ook dat gezondheidseffecten foutief aan een statistisch significante maar toxicologisch onschadelijke polluent toegeschreven worden.

Een eerste doelstelling van dit doctoraat is om aan te tonen dat, door gebruik te maken van nieuwe en innovatieve meettoestellen, het meten van persoonlijke blootstelling haalbaar is en nuttige informatie oplevert. Daarnaast werd reeds aangetoond dat activiteiten gebaseerde modellen veelbelovend zijn bij het modelleren van de blootstelling van een populatie; in dit doctoraat zal deze modelketen fundamenteel gewijzigd worden voor het berekenen van persoonlijke blootstelling.

Deze nieuw ontwikkelde methoden worden toegepast voor de polluent black carbon (BC), een component van fijn stof en in Vlaanderen hoofdzakelijk afkomstig van verkeer. Gedurende de laatste decennia zijn BC concentraties gedaald in geïndustrialiseerde landen, maar de lucht is toch nog steeds matig tot sterk vervuild. Meerdere studies tonen aan dat BC verantwoordelijk is voor gezondheidseffecten bij de mens, en BC speelt ook een belangrijke rol in de

opwarming van de aarde, mede daardoor is de polluent opgenomen in verscheidene actuele beleidsdocumenten.

Materialen en methoden

Persoonlijke blootstelling aan luchtvervuiling kan bepaald worden door middel van indirecte blootstellingsmodellen of door een directe aanpak (luchtkwaliteitsmetingen). Het nieuwe activiteiten gebaseerde model voor Vlaanderen FEATHERS wordt gebruikt om te modelleren waar en wanneer mensen activiteiten uitvoeren (vb. werken, winkelen, slapen, zich verplaatsen). Land use regression (LUR) modellen worden opgesteld om de luchtkwaliteit met een hoge ruimtelijke en tijds-resolutie in kaart te brengen. In een LUR model worden statistische associaties gezocht tussen potentiële predictorvariabelen en metingen, in dit geval op 63 vaste locaties, als basis voor het voorspellen van concentraties op locaties waar niet gemeten werd. LUR voorspellingen worden vervolgens gecombineerd met een indoor/outdoor factor voor blootstelling in indoor omgevingen. Blootstelling tijdens verplaatsingen wordt ingeschat door een speciaal daartoe ontwikkeld model dat rekening houdt met transportmiddel, tijdstip van de verplaatsing en urbanisatiegraad. De gehele modelketen wordt gevalideerd aan de hand van persoonlijke metingen van 62 vrijwilligers. Deze meetcampagne omvat het persoonlijk meten van BC met micro-aethalometers, en het verzamelen van dagboekjes en GPS logs op een elektronisch platform gedurende 7 opeenvolgende dagen. Dit resulteert in meer dan 10000 uren data, en de registratie van meer dan 1500 verplaatsingen.

Resultaten en conclusies

Uit de persoonlijke metingen is gebleken dat blootstelling aan BC tot wel 30% kan verschillen tussen partners die op eenzelfde adres wonen. Meestal zijn huisvrouwen of huismannen blootgesteld aan lagere concentraties in vergelijking met voltijds werkende personen, vooral door het verschil in reistijd en de verhoogde concentraties waargenomen tijdens verplaatsingen. De deelnemers aan de meetcampagne brachten gemiddeld 6% van hun tijd door in transport, maar dit zorgt wel voor 21% van hun blootstelling aan BC, en zelfs voor 30% van de dagelijkse ingeademde dosis. Helaas bleek reistijd geen goede voorspeller voor geaccumuleerde persoonlijke blootstelling omdat vele factoren de blootstelling in transport mee bepalen. De blootstelling is het hoogst in gemotoriseerde modi (auto, bus, tram / metro), en het laagst voor fietsers, voetgangers en in treinen; blootstelling wordt belangrijker voor fietsers en voetgangers als er rekening wordt gehouden met de ingeademde dosis. In een voertuig zijn de concentraties het hoogst op snelwegen en in een stedelijke omgeving, op spitsuren en op weekdagen, bij snelheden <30 km/h en >80 km/h, en de concentraties nemen toe wanneer men rijdt op wegen met druk verkeer. Deze laatste factor bleek de belangrijkste verklarende variabele voor blootstelling in een voertuig, samen met tijdstip van de verplaatsing en urbanisatiegraad. Voor fietsers en voetgangers wordt blootstelling vooral verklaard door tijdstip van de verplaatsing en urbanisatiegraad.

Persoonlijke blootstelling wordt gemodelleerd door gebruik te maken van het activiteiten gebaseerde model FEATHERS, LUR modellen voor elk uur van de dag, een indoor luchtkwaliteitsmodel, en een model dat blootstelling in transport voorspelt. Deze componenten worden gecombineerd in AB²C (Activiteiten geBaseerde modelketen voor het bepalen van blootstelling aan BC). Het opstellen van onafhankelijke uurlijkse LUR modellen resulteerde in R^2 waarden die meestal kleiner waren dan de R^2 van het jaargemiddelde model (R^2=0.77), namelijk tussen 0.07 en 0.8. Op weekdagen tussen 6u in de ochtend en 22u 's avonds benaderde de R^2 de jaargemiddelde R^2. Zelfs al worden modellen voor opeenvolgende uren onafhankelijk van elkaar ontwikkeld, toch bleken gelijkaardige variabelen significant te zijn. Het gebruik van dynamische variabelen in plaats van statische, dit wil zeggen verkeersstromen en bevolkingsdichtheden voorspeld door FEATHERS voor elk uur van de dag, bleek de performantie van de LUR modellen niet te verhogen.

De AB²C modelketen wordt gevalideerd met de tijds-activiteitenpatronen en de BC concentraties zoals gemeten bij 62 vrijwilligers. Voor elke deelnemer aan de meetcampagne wordt een synthetische populatie van 100 model-agents per dag samengesteld waarbij alle agents dezelfde kenmerken hebben als een real-life agent. Het AB²C model berekent vervolgens voor elk individu een verdeling van mogelijke blootstellingen. De inschatting van de gemiddelde persoonlijke blootstelling met AB²C blijkt iets nauwkeuriger in vergelijking met buitenluchtconcentraties zoals die voorspeld worden voor de woonzone. De meerwaarde van de AB²C modelketen ligt momenteel voornamelijk in de mogelijkheid voor het detecteren van herhaaldelijke korte termijn piek bloostellingen, eerder dan in het modelleren van een gemiddelde blootstelling.

In dit doctoraat wordt er aangetoond wat de mogelijkheden, de voordelen en de beperkingen zijn van gepersonaliseerde blootstellingsmonitoring. Er is een activiteiten gebaseerde modelketen ontwikkeld die persoonlijke bloostelling berekent, en die kan gebruikt worden voor het bepalen van de blootstelling van individuen in een populatie. Hiervoor zijn er submodellen ontwikkeld die op zichzelf uniek zijn, maar die ook gekoppeld zijn om te resulteren in het innovatieve persoonlijke blootstellingsmodel AB²C.

ABBREVIATIONS

AB^2C	Activity-Based modeling framework for Black Carbon exposure assessment
BC	Black Carbon
BS	Black Smoke
CH_4	Methane
CHD	Coronary Heart Disease
CI	Confidence Interval
CO	Carbon Monoxide
CO_2	Carbon Dioxide
DPM	Diesel Particulate Matter
EC	Elemental Carbon
EEA	European Environment Agency
ETS	Environmental Tobacco Smoke
EU	European Union
GIS	Geographic Information System
GPS	Global Positioning System
HIA	Health Impact Assessment
I/O-ratio	Indoor/Oudoor ratio
iF	Intake Fraction
IQR	Interquartile Range
IR	Inhalation Rate
LOOCV	Leave-One-Out Cross-Validation
LUR	Land Use Regression
NO	Nitrogen Monoxide
NO_2	Nitrogen Dioxide
NOx	Nitrogen Oxides
O_3	Ozone
OC	Organic Carbon
PDA	Personal Digital Assistant
PM_{10}	Particulate Matter with an aerodynamic diameter of 10 µm or less
$PM_{2.5}$	Particulate Matter with an aerodynamic diameter of 2.5 µm or less

$PM_{2.5}$abs	$PM_{2.5}$ absorbance
PM_{coarse}	Coarse Particulate Matter, difference between PM_{10} and $PM_{2.5}$
r	Correlation Coefficient
RMSE	Root Mean Square Error
SO_2	Sulfur Dioxide
TAZ	Traffic Analysis Zone
U.S. EPA	United States Environmental Protection Agency
UFP	Ultrafine Particles ($PM_{0.1}$)
VIF	Variance Inflation Factor
VOC	Volatile Organic Compounds
WHO	World Health Organization
WKDY	Weekday
WKND	Weekend

CONTENTS

1. GENERAL INTRODUCTION

1.1 PROBLEM STATEMENT

Evidence is growing that a myriad of health effects, ranging from respiratory to cardiovascular and neurological effects, are causally linked to exposure to air pollution. Annually an estimated number of 2 million premature deaths can be attributed to air pollution worldwide. In recent years, society has become increasingly sensitive to individual risks, and thus data on the exposure to air pollution needs to be personalized.

According to the National Research Council, Committee on Human and Environmental Exposure Science in the 21st Century (reported in Lioy and Smith (2013)), exposure science can be defined as: "the collection and analysis of quantitative and qualitative information needed to understand the nature of contact between receptors and physical, chemical, or biologic stressors. Exposure science strives to create a narrative that captures the spatial and temporal dimensions of exposure events with respect to acute and long-term effects on human populations and ecosystems". Traditionally exposure of the population to air pollution has been calculated by ignoring spatial and temporal dimensions of exposure. More complex models, taking into account population movement and/or changing air quality, are challenging with respect to data requirements and are therefore rare. Obtaining personal air pollution exposure measurements has been hampered by the cost and complexity of the analyzing equipment.

The association between health effects and specific sources of air pollution or specific air pollutants remains a source of scientific controversy and uncertainty for policy makers (Knol et al., 2009). By using novel methods and devices, it will be possible to estimate exposure to specific air pollutants more accurately. In the future this will reduce exposure misclassification in epidemiological studies, and the linkages between source – stressor – receptor – health outcome will be established with more certainty. As a result, public health and traffic policies aimed at reducing risks of air pollution will become more cost-efficient.

1.2 RESEARCH OBJECTIVES

The overall objective of this dissertation is to develop and validate a chain of models that is able to estimate the accumulated exposure of an individual in the region of Flanders, Belgium.

At the start of the project, the hypothesis was formulated that visiting certain microenvironments is the most important determinant of personal exposure, and that the movement between microenvironments differentiates exposure between people living in the same geographical zone. To test this hypothesis two complementary methodologies are used: modeling and measuring.

- The newly developed Flemish activity-based model FEATHERS is used to model the activities that people perform (e.g. work, shopping, sports …) as well as the times and locations;
- Newly developed portable monitors for air pollution are being deployed in a subgroup of the population to measure personal exposure to traffic-related air pollution.

To reach the overall objective, four specific goals are defined:

1. to demonstrate the feasibility of personal monitoring to assess exposure to air pollution;
2. to develop and demonstrate the data-mining capabilities necessary to handle and analyze the datasets derived from mobile measurements;
3. to produce hourly concentration maps using land use regression techniques;
4. to take full advantage of the FEATHERS activity-based model so that it can predict personal exposure to air pollutants at the population level.

Existing and newly developed models will be integrated in a modeling framework, with FEATHERS being the base model, to assess exposure of individual agents or groups of agents. The framework will be validated with personal measurements.
The developed methodologies are applied for the air pollutant black carbon (BC), which is identified as an important proxy for traffic-related pollution. It is shown that BC has important health effects in humans, and it is a potent climate

warmer (Bond et al., 2013; Janssen et al., 2011). Because BC has steep gradients when moving away from roads (Karner et al., 2010; Zhu et al., 2002), detailed concentration maps are advantageous, and taking population mobility into account is highly relevant. BC can be measured both in a mobile and in a fixed setting using small monitors (AethLabs, 2011).

The developed modeling framework can be used to evaluate the impact of policy measures on personal and population exposure to air pollution. This model will enable the assessment of general traffic policy measures on transport, concentrations, exposure, and human health. Next to transportation measures, activity-based models are able to calculate the effects of certain scenarios with no obvious relation to transport or air quality. Institutional changes (e.g., changing working hours, changing shop opening hours, congestion charging) or demographical changes (e.g., aging of the population, changing percentage of part-time workers, more one-adult households) can be assessed with an activity-based model; all are evolutions that are relevant to current policy.

1.3 OUTLINE OF THE THESIS

This first introductory chapter gives a rough description of the problem and states the research aims.

In chapter 2, current knowledge in the broader field of air pollution exposure assessment is summarized. This chapter provides details on exposure to air pollution in general, and then focuses on direct measurement and indirect modeling of exposure. Background information is presented on the techniques and models that will be used in chapters 3 and 4 of the dissertation. Chapter 2 ends with a succinct overview of recent literature on the air pollutant black carbon (BC).

Chapter 3 describes the results of a personal monitoring campaign. Volunteers were instructed to carry a device measuring BC and an electronic diary with GPS for 7 consecutive days, while performing their everyday activities. Part 3.1 explores the impact of time-activity patterns on personal exposure. Results are presented from a pilot study in 8 couples and in one season. Part 3.2 elaborates on this first paper, with an expansion of the number of participants up to 62 and with measurements in two seasons. This chapter focuses on exposure while traveling and calculates inhaled doses in different transport modes. Part 3.3 then investigates trip and road characteristics influencing in-vehicle concentrations and concentrations while cycling or walking.

The aim of chapter 4 is to model the accumulated exposure of an individual to BC. Therefore BC measurements are carried out in the study area, both on urban and regional fixed locations in two campaigns. The land use regression technique is used to calculate multiple regression models using measurements from both campaigns; results are presented in part 4.1. Part 4.2 builds on these measurements, but calculates land use regression models with an hourly temporal resolution. As a final step, the hourly concentration maps are combined with other models in the AB²C (Activity-Based modeling framework for Black Carbon exposure assessment) modeling framework to predict personal exposure in part 4.3. The model is evaluated by comparing the model estimates with revealed exposures from chapter 3.

The last chapter, chapter 5, discusses the contributions of this dissertation to the state-of-the-science. The most important findings and knowledge gaps are discussed as well. The chapter concludes with recommendations for future health studies.

FIGURE 1 gives a schematic overview of the outline of the dissertation. Chapters 3 and 4 are largely based on published or submitted peer-reviewed papers as detailed below.

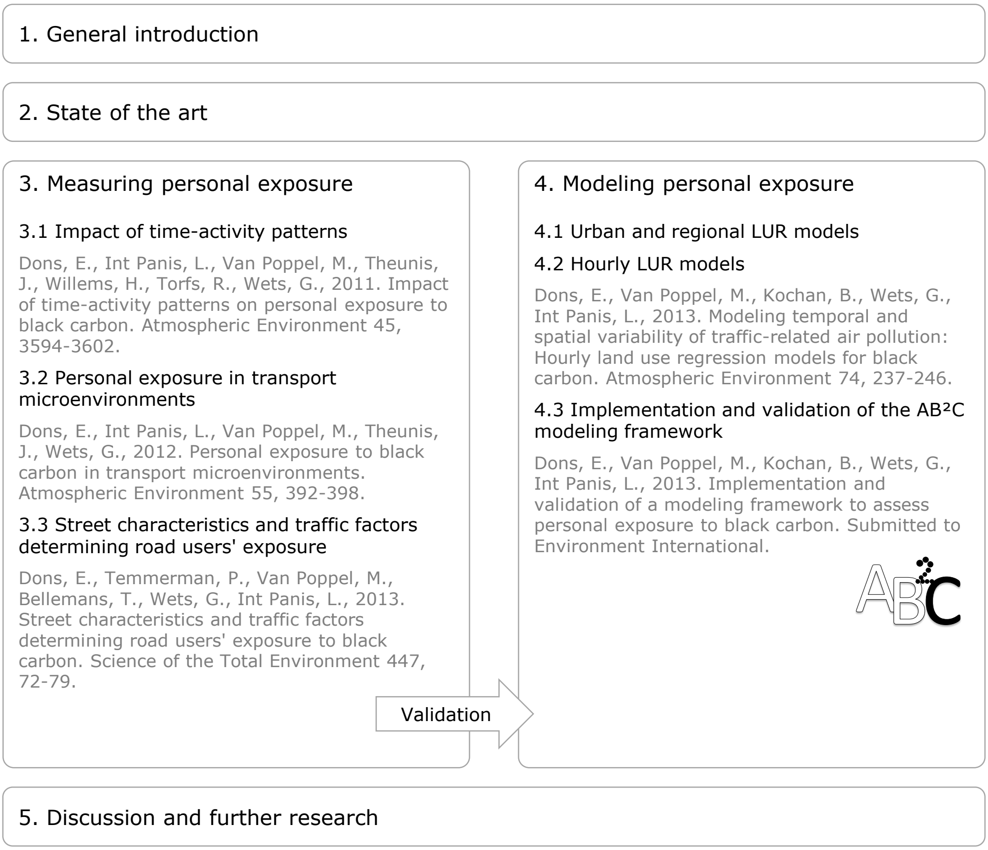


FIGURE 1: Outline of the dissertation with references to the relevant chapters in this book and published or submitted peer-reviewed papers.

2. STATE OF THE ART

2.1 PERSONAL EXPOSURE TO AIR POLLUTION

Worldwide, air pollution causes an estimated 2 million early deaths each year (WHO, 2013). Air quality and the effects of typical ambient air pollution concentrations remain an important public health risk. Mortality, cancer, cardiovascular symptoms, respiratory and neurological effects have all been linked to air quality (Brook et al., 2010; Calderon-Garciduenas et al., 2008; Pope and Dockery, 2006). Improving air quality, both in the light of climate change mitigation and for the prevention of health effects, is a global challenge.

2.1.1 AIR POLLUTION: THE BIGGER PICTURE

An integrated approach to describe environmental problems is the DPSIR framework, based on a model of the OECD (EEA, 1999; OECD, 1993). The model describes the chain of environmental disturbances and causally links human activities and damage to the environment. Driving forces (D) like performing out-of-home activities, transport and industrial activities lead to environmental pressures (P) that degrade the state (S) of the environment. This state, e.g. the air quality, has an impact (I) on human health and the natural environment which provokes society to carry out a response (R).

Being in transport is most of the time not an aim on itself, but this is driven by activities individuals wish or need to perform. The distance people need to travel depends a.o. on the spatial planning in a region; the combination of the demand for activities and the supply of services in a region can be described as the 'trip market'. When making a trip from location *A* to location *B* at time *t*, one can choose from a number of different transport modes and services (transportation market). The resulting trips with each transport mode can then be assigned to available infrastructure, e.g. the road network, bus lines (traffic market).

In FIGURE 2, the DPSIR framework and the 3-markets model are integrated in one framework that describes the chain of environmental disturbances from activities to impact and responses.

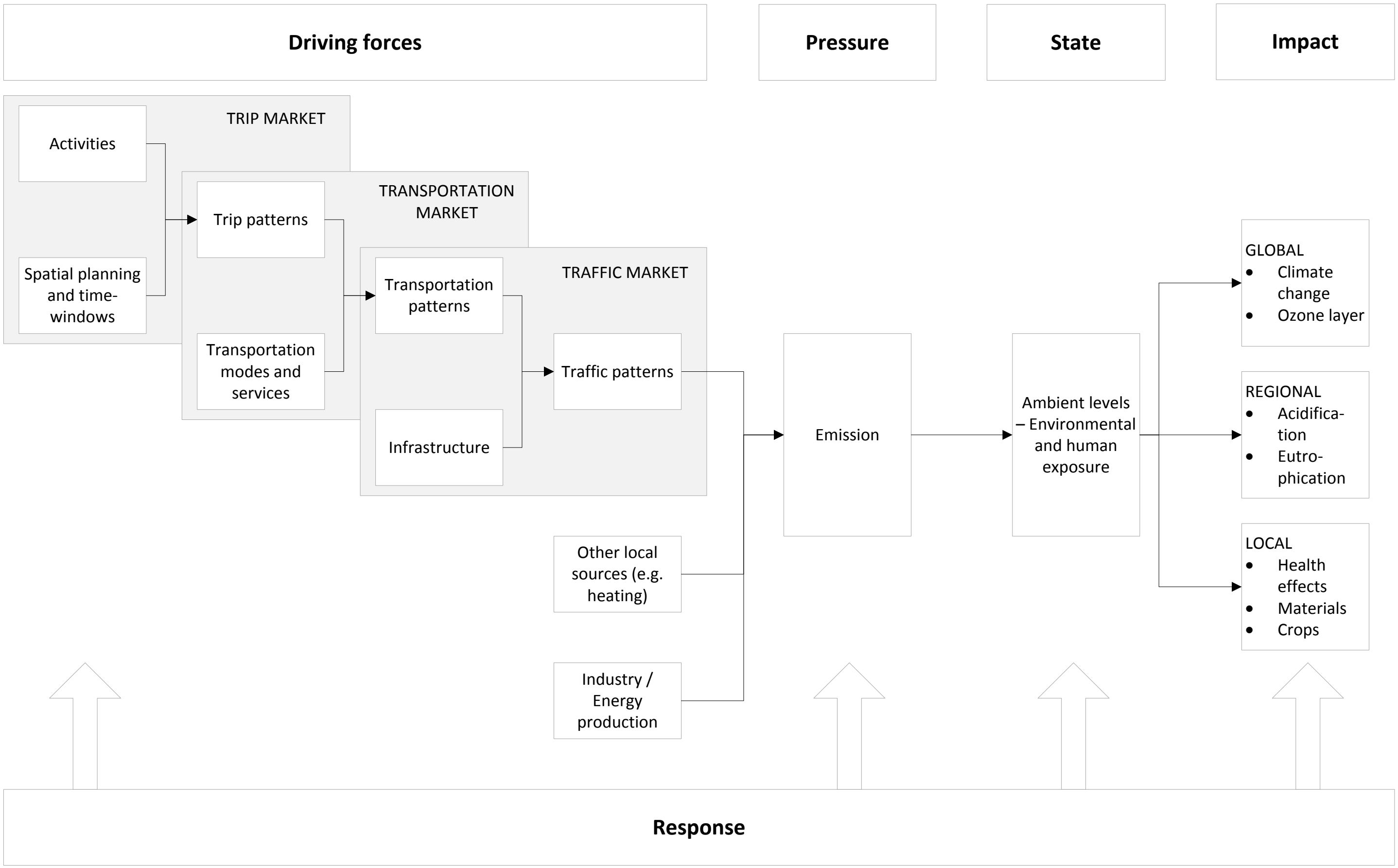


FIGURE 2: The DPSIR concept applied to air pollution, and extended with the 3-markets model to formulate transportation policy (adapted from (Jensen, 1999))

One of the most important air pollution impacts is the impact on health. The process through which air pollution impacts human health, the 'environmental pathway', is presented in FIGURE 3. Upstream factors, e.g. land use, transport or energy policy; human activities including traffic, heating, and industrial activities; and natural sources of pollutants; all contribute to emissions. Emissions from point sources (smokestacks), line sources (roads) or area sources (cities) will disperse, transform, interact with other pollutants, and eventually disappear from the atmosphere. Dispersion of emitted pollutants will result in different levels of air pollution concentrations on every location, both indoors and outdoors: the 'environmental intensity'. Individuals or receptors move through space and time and are exposed to local concentrations while performing activities. People are not only 'externally exposed', but pollutants can also enter the body mainly through inhalation in the case of air pollutants, resulting in an inhaled dose. A causal link is established between exposure to air pollution and certain health effects; for other health effects evidence is suggestive (HEI, 2010). The separation of time-activity into its own category emphasizes its importance in exposure science: exposure should be assessed by combining data on environmental intensity and time-activity profiles (Lioy and Smith, 2013). Feedback loop (1) shows that an outcome experienced by an individual could spread to others. Feedback loop (2) includes how health outcomes can alter activities and behaviors among individuals, for example susceptible populations are discouraged to exercise during ozone peaks.

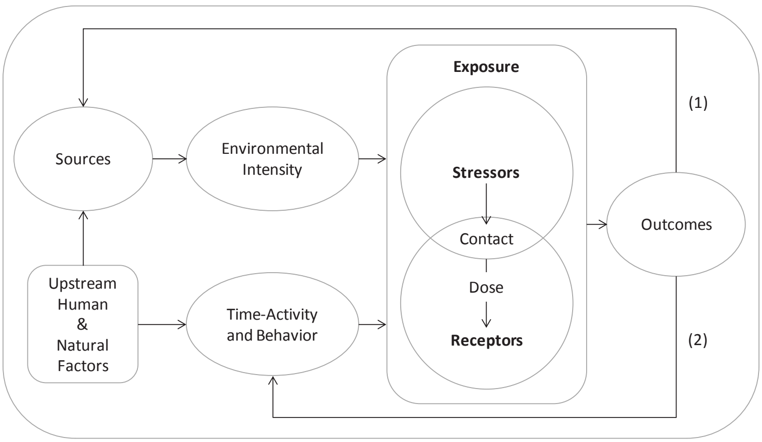


FIGURE 3: Causal network from activities to health outcomes via air pollution (Lioy and Smith, 2013)

2.1.2 AIR POLLUTION: EMISSIONS, DISPERSION AND LIMIT VALUES

Air quality on a particular location at a certain point in time is the result of a combination of emissions and dispersion of the emissions. Gases or particles are put into the air by various sources, both from natural and from anthropogenic origin, and they are physically and chemically diverse and complex. The most important natural sources include sea spray, dust storms, volcanic eruptions, grassland and forest fires (EEA, 2012). But human activities are the main cause of poor air quality: traffic, industry, or biomass burning, are among the most important contributors.

Atmospheric dispersion describes the transport of emitted pollutants in the lower atmosphere. Dispersion conditions are influenced mainly by local circumstances (e.g. street canyons versus open areas), weather conditions, and orography (elevation). Pollutants often transform during transport through coagulation processes, nucleation, deposition, condensation, or (photo)chemical reactions. Air pollution dispersion models describe these phenomena in mathematical functions; examples of widely used dispersion models are AERMOD, ADMS, CALINE4, IFDM and CALPUFF. Most of these models combine data on emissions (a.o. traffic emissions that depend on the volume and composition of traffic) with data on weather conditions (such as wind speed, wind direction and mixing height) to simulate dispersion processes.

In developed countries, motor vehicles represent a major source of air pollutants that have a substantial impact on ambient air pollution, indoor air, and personal exposures. Traffic-related air-pollution is a complex mix of components and much is unknown about the toxicity of the different components (Laumbach and Kipen, 2012). Traffic emissions contribute to both primary air pollutant concentrations (BC, NOx, CO, benzene, UFP, PM, VOC) that are emitted directly from tailpipes, and to secondary pollutant concentrations (NO_2, O_3, secondary (in)organic aerosols) that are formed in the atmosphere from precursors (Erisman and Schaap, 2004; HEI, 2010; Int Panis, 2008; Marshall et al., 2005). Primary exhaust emissions in Europe are regulated via the EURO emission standards (IIASA, 2012). Non-exhaust emissions (PM_{coarse}, Cu, Zn, Fe) include brake and tire wear, resuspended dust and road abrasion; non-exhaust emissions are currently beyond legislation (IIASA, 2012).

When estimating the impact of traffic on ambient concentrations and exposure, most studies focus on single compounds, referred to as carriers or markers, although specific health effects may not be caused by just one component but by the collective attributes of the 'cocktail' (HEI, 2010). Alternatively, direct measures of traffic (traffic intensity on the nearest road, road length in buffers, etc.) are used to quantify exposure to traffic-related air pollution.

The contribution of traffic to local air quality is largest in cities at locations near major roads. Concentrations measured can be decomposed in a transnational part (emissions from other countries), a national part (national emissions), an urban contribution (emissions from local industries, households, urban traffic), and a contribution of local sources (traffic in the direct vicinity). Depending on the pollutant, the location, and the time period, the relative contribution from each part can be very different.

To limit the negative effects associated with air pollution, legally-binding limit values are formulated by different authorities. Primarily, emission ceilings regulate maximum emissions in different sectors and countries. In addition, air pollutant concentrations cannot exceed predefined limits (TABLE 1). Air quality standards are not globally harmonized, e.g. NO_2 limit values are much stricter in Europe than in the United States, whereas for $PM_{2.5}$ the opposite is true (in December 2012, U.S. EPA even strengthened the $PM_{2.5}$ annual standard to 12 µg/m³). The WHO has formulated its own standards based on scientific evidence in response to the real and global threat to public health (WHO, 2005). When countries or regions set or revise ambient air quality standards, air quality monitoring data are considered most frequently, followed by existing standards in other countries, environmental epidemiology studies, and the WHO guidelines (Vahlsing and Smith, 2012).
Air quality standards are intended to protect people to exposures above a certain threshold; although there is no evidence of a level below which there is

no risk for adverse health effects (Lioy and Smith, 2013; U.S.EPA, 2012a). Compliance with the standards is controlled by fixed air quality monitoring stations continuously measuring a range of air pollutants. This includes traffic, urban and regional sites, but also industrial sites. As people are not always near a monitor representative for their exposure, it does happen that people are exposed to excessive concentrations. The EU has introduced the Exposure Concentration Obligation for $PM_{2.5}$: a target solely based on concentrations on urban background sites better reflecting population exposure (limit value of 20 µg/m³ to be met in 2015).

TABLE 1: Air quality standards in Europe and the US, and guideline values from the WHO

	EU	US	WHO
NO_2	200 µg/m³ (1h mean, 18 times/year) 40 µg/m³ (annual mean)	191 µg/m³ (1h mean, 98th percentile) 100 µg/m³ (annual mean)	200 µg/m³ (1h mean) 40 µg/m³ (annual mean)
PM_{10}	50 µg/m³ (1day mean, 35 times/year) 40 µg/m³ (annual mean)	150 µg/m³ (1day mean, 1 time/year) 50 µg/m³ (annual mean)	50 µg/m³ (1day mean) 20 µg/m³ (annual mean)
$PM_{2.5}$	25 µg/m³ (annual mean)[a]	35 µg/m³ (1day mean, 98th percentile) 12 µg/m³ (annual mean)	25 µg/m³ (1day mean) 10 µg/m³ (annual mean)
SO_2	350 µg/m³ (1h mean, 24 times/year) 125 µg/m³ (1day mean, 3 times/year)	366 µg/m³ (1day mean) 78 µg/m³ (annual mean)	20 µg/m³ (1day mean)
O_3	120 µg/m³ (8h mean, 25 days/year)[a]	157 µg/m³ (8h mean)	100 µg/m³ (8h mean)

[a] This is a target value rather than a limit value

2.1.3 PERSONAL EXPOSURE AND DOSE

Personal exposure can be defined as the true exposure experienced by individuals. When individual *i* is at location *x,y,z*, then the concentration on that location *x,y,z* is his personal exposure for a given time period. But when the individual travels from location A to location B, the exposure is determined by the time-weighted concentrations on both locations, increased with the exposure while in transport (WHO, 1999). The exposure of a complete population is the sum of all individual exposures.

Formally, personal exposure to air pollution can be presented as a multiplication of concentrations in microenvironment *i* (C_i) with the time spent in that microenvironment ($time_i$) (Duan, 1991; Klepeis, 1999, 2006; Nethery et al., 2008b; Ott, 1982; Sexton and Ryan, 1988). Integrated personal exposure is then the sum of the products of concentrations and the time spent in the respective microenvironments.

$$Personal\ Exposure = \sum_{i=0}^{all\ i\ microenvironments} C_i \times time_i$$

The latter formula can be transformed into an algebraic function with $C_i(t,x,y,z)$ being the concentration as experienced by individual *i* on time *t* on a location with geographical coordinates [x,y,z]. The start- and endpoint t_1 and t_2 mark the exposure episode (Klepeis, 2006).

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

In a figure, the integrated cumulative exposure can be presented as follows (exposure episode is marked with *0* and t_a):

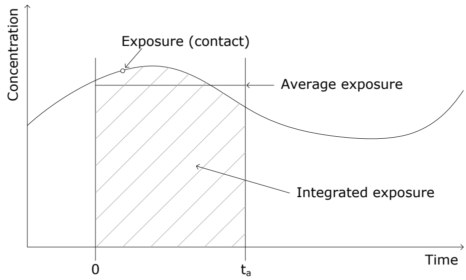


FIGURE 4: Definition of exposure (Monn, 2001)

The formula can then be adapted again so that specific, and thus discrete, microenvironments are regarded as spatial variable, rather than the continuous space (Klepeis, 2006). C_{ij} is the concentration experienced by the receptor in the discrete microenvironment *j* at a particular point in time *t* over the time interval defined by [t_{j1};t_{j2}]. This can again be summed over all m microenvironments.

$$E_i = \sum_{j=1}^{m} (\int_{t_{j1}}^{t_{j2}} C_{ij}(t)dt)$$

Personal exposure levels can be estimated directly or indirectly (Piechocki-Minguy et al., 2006). The direct approach applies portable measurement devices that can be worn by individuals for a short time ('external exposure'). In addition, exposure or dose biomarkers are defined, determining past exposures of individuals to air pollutants ('internal exposure'). Unlike e.g. exposure to ETS that can be determined from cotinine levels in blood, urine or saliva (Benowitz, 1999), few if any suitable biomarkers of exposure to traffic related air pollution exist (e.g. airway macrophage carbon in sputum (Nwokoro et al., 2012)). On the other hand, indirect approaches use various models that reconstruct exposure from time-activity databases and concentration maps. Because it is not realistic to use the direct approach in larger populations (air quality devices are expensive and cheaper sensors are not yet reliable, dose biomarkers use invasive and labor-intensive techniques), models should be applied to estimate personal exposure in larger cohorts.

In the past, human exposure to air pollution was estimated by using concentrations measured at a few fixed air quality monitoring stations. Concentrations measured at the monitoring station(s) nearest to a person's home are assumed to be representative of exposure of an individual (Fenske, 2010; Kaur et al., 2007; Lepeule et al., 2010; Park et al., 2010; Peters et al., 2009; Sarnat et al., 2010). Unfortunately, the spatial density of fixed monitors is insufficient and the position of the monitoring sites is often not representative for the exposure of a population (Briggs et al., 2000). Personal exposure of individuals does not only depend on ambient concentrations, but also on the concentrations encountered in specific microenvironments (including indoor sources), and on the time-activity patterns of individuals (Boudet et al., 2001; Brown et al., 2008; Jensen, 1999; McKone et al., 2009). The correlation between personal exposure and concentrations measured at fixed sites is mostly low for pollutants with high spatiotemporal variability like NO_x or BC, but can be better for $PM_{2.5}$ or PM_{10} (Baxter et al., 2007; Sarnat et al., 2006), although significant differences exist between different studies (Avery et al., 2010). Some studies find that personal exposure is higher than ambient concentrations measured at fixed sites (Avery et al., 2010; Broich et al., 2012; Brown et al., 2009b; Monn et al., 1997). Monn et al. (1997) and Wallace et al. (2006) refer to this phenomenon as the ‘personal cloud’; the origin of this cloud is however unknown. Jerrett et al. (2005a) postulate that personal exposure measurements lead to lower exposure values compared to measurements at fixed sites because indoor concentrations tend to be lower. As people spent 80 to 90% of their time in indoor microenvironments (Klepeis, 2006), personal exposure tends to be better correlated to indoor concentrations compared to ambient concentrations (Brown et al., 2008; Koistinen et al., 2001; Piechocki-Minguy et al., 2006).

The large variation in correlations between personal, ambient, and indoor concentrations, demonstrates that exposure misclassification can be significant when only using concentrations on fixed monitoring sites. If exposure is considered on the aggregated level of a population, these differences are probably averaged out, but exposure as experienced by individuals, including related health effects, will be strongly impacted by variations in personal non-ambient concentrations (McKone et al., 2009). Only recently, Setton et al. (2011) and Baxter et al. (2013) confirmed that time away from home and time in transport are significant effect modifiers when determining health effects associated with exposure to air pollution. Von Klot et al. (2011) found an association between personal BC exposure and acute myocardial infarction, but not with ambient $PM_{2.5}$ concentrations, suggesting that it is important to include out-of-home activities. The relationship between personal, ambient and indoor concentrations cannot be determined unambiguously, and depends on the individual, his/her time-activity pattern and the environment.

Rather than focusing on exposure, the concept of ‘dose’ is important when considering health effects associated with air pollution. An individual may be exposed to a pollutant, but that pollutant does not necessarily enter the body of the individual (Ott, 1982). A dose only occurs when a pollutant crosses a physical boundary, i.e. when a person inhales or ingests the air pollutant. As an example: two individuals are present at the same athletics field, one person is running and the other one is watching, their exposure will be nearly the same as they are within meters of each other, but the runner inhales more particles due to his faster and deeper breathing. The amount of inhaled air depends partly on personal characteristics: e.g. gender, age, and weight (Oravisjärvi et al., 2011);

the other part can be explained by performing (physical) activities (Marshall et al., 2006; McConnell et al., 2010). The importance of breathing rates while traveling is shown by Int Panis et al. (2010) and Zuurbier et al. (2010).

The amount of air moving through the lungs per time unit is expressed as minute volume (l/min). The amount of inhaled air during a day is estimated as a time-weighted average at different levels of effort. The 24-h inhalation rate with *n* different levels of effort can be determined as (Allan and Richardson, 1998):

$$IR = \frac{1}{10^3}\sum_{t=1}^{n} t_i \times V_i$$

with IR = the 24-hour inhalation rate (m^3/day);
t_i = the amount of time spent at activity level i (min/day);
V_i = the minute volume at activity level i (l/min);
10^3 = a conversion factor (L/m^3).

Several authors or institutions have determined breathing rates for different activities (Int Panis et al., 2010; Layton, 1993; U.S.EPA, 2011; Zuurbier et al., 2010). In a paper of Allan and Richardson (1998), 5 different activity levels are defined with corresponding minute ventilation (TABLE 2).

- Activity Level 1 - resting (e.g. sleeping, resting, watching TV, reading);
- Activity Level 2 - very light activity (e.g. services, working);
- Activity Level 3 - light activity (e.g. home activities, shopping, working);
- Activity Level 4 - light to moderate activity (e.g. gardening, dancing);
- Activity Level 5 - moderate to heavy activity (e.g. sports, walking).

TABLE 2: Overview of minute volume assumptions in l/min for adults and seniors (Allan and Richardson, 1998)

Activity Level	Description	Male adult[a,b]	Female adult[a,b]	Male senior[a,b]	Female senior[a,b]
1	Resting	8.3 ± 2.8	7.5 ± 2.5	8.2 ± 2.2	6.8 ± 1.9
2	Very light activity	10.5 ± 3.3	12.5 ± 3.9	10.7 ± 3.1	10.1 ± 3.0
3	Light activity	16.1 ± 4.1	13.0 ± 3.3	16.2 ± 3.8	13.2 ± 3.1
4	Light to moderate activity	30.2 ± 4.9	23.2 ± 3.8	31.1 ± 6.0	24.0 ± 4.6
5	Moderate to heavy activity	49.2 ± 10.6	39.8 ± 8.6	59.3 ± 10.0	49.2 ± 8.3

[a] All lognormal distributions, except for activity level 5 (normal distribution)
[b] Adults (20-59 years); seniors (60+ years)

Deposition models, like the models from the International Commission on Radiological Protection (ICRP) and the National Council on Radiation Protection and Measurements (NCRP), can be used to estimate in which part of the lung certain particles deposit. These models estimate the total and the regional lung deposition of aerosols of different sizes and under different breathing rates. For example, in a study of Oravisjärvi et al. (2011) the deposition of traffic-related particles from diesel buses in the lungs is simulated. Buonanno et al. (2011) determine the daily tracheobronchial and alveolar dose of ultrafine particles for a synthetic population in Italy. The exposure and the inhaled dose are determined by applying time-activity patterns and corresponding ventilation rates.

The origin of inhaled pollutants is important when determining health effects of certain levels of air pollution. Particulate matter emissions from some natural sources are thought to be less damaging than traffic-related air pollution (EEA, 2012). This can be explained by differences in toxicity (see next paragraph) and because of differences in the intake fraction (iF), which is defined as the ratio between the total amount of inhaled particles and the total amount emitted. An iF can be considered as a simplified combination of emission, dispersion and exposure models in one number. From this ratio it appeared that only a small portion of the particles emitted is in fact inhaled by an individual. The iF's of 3646 cities from all over the world were reported by Apte et al. (2012); eleven Belgian cities have an average iF of 13 ppm (1 ppm = 1 g inhaled / t emitted). This ratio is not the same for particles from different sources: emissions from road traffic are more often inhaled than particles from airplanes (Tainio et al., 2009). The health impact of a pollutant can be expressed in the following formula:

$$Health\ Impact = Total\ Mass\ Emitted \times Intake\ Fraction \times Toxicity$$

Pollutants with an important health impact have either an elevated iF or a higher toxicity or both.

2.1.4 HEALTH EFFECTS ASSOCIATED WITH EXPOSURE TO AIR POLLUTION

Health effects that are linked to exposure to air pollution can be divided into short term acute effects and long term effects. Short term exposures are marked by a short episode of high concentrations, whereas long term health effects are a consequence of elevated concentrations over a longer time span.

2.1.4.1 Short term health

The earliest studies that evaluated short term health effects focused on severe air pollution episodes and mortality; examples are the Meuse Valley fog of 1930 and the London fog of 1952 that caused hundreds of deaths (Bell and Davis, 2001; Nemery et al., 2001). More recently, in a review paper from Pope and Dockery (2006) different US and European multicity studies are listed that all give similar results: a more or less linear rise in the number of deaths with increasing PM_{10} and black smoke concentrations. In the APHEA2 project, short-term health effects of ambient particles on total nonaccidental mortality from 29 European cities were reported (Katsouyanni et al., 2003). It was found that PM air pollution was significantly associated with both respiratory and cardiovascular daily mortality counts. Next to mortality effects, also effects on morbidity are observed. Nawrot et al. (2011) did a literature review on triggers of myocardial infarction: air pollution (a difference in PM_{10} concentrations of 30 µg/m³) is one of the most important triggers, next to participation in traffic, physical activity and alcohol. From a study of Peters et al. (2004) it appeared that the risk of myocardial infarction increases when a patient was in transport the hour before the incident (odds ratio = 2.92); the risk is largest when traveling by bike (odds ratio = 3.94) or by severe exertion (odds ratio = 6.38). The relative contribution of risk factors such as stress, traffic-related air pollution, or noise is impossible to determine from this study. Brook et al. (2010) summarized current knowledge on particulate matter exposure and cardiovascular disease: they concluded that exposure to $PM_{2.5}$ over a few hours to weeks can trigger cardiovascular disease-related mortality and nonfatal events, and that this relationship is causal. Mills et al. (2007) found ischemic and thrombotic effects after short term peak exposures to diesel exhaust in 20 patients with coronary heart disease. This study in a controlled environment with concentrations several times larger than ambient levels was important as it proved biological plausibility of acute health effects associated with air pollution. A similar study of Lucking et al. (2011) found that the presence of a particle trap on diesel engines prevents several adverse cardiovascular effects that did show when volunteers were exposed to unfiltered diesel exhaust (although NO_2 concentrations were a factor 5 higher in filtered air). McCreanor et al. (2007) did a randomized crossover study in patients with asthma; clear respiratory effects were shown to be caused by exposure to diesel traffic.

In Flanders, a relationship was found between daily mortality and PM_{10} concentrations, especially during summer (Nawrot et al., 2007). From the same study, it was estimated that 630 premature deaths could be avoided when PM_{10} did not exceed 20 µg/m³; this effect is largest in summer.

2.1.4.2 Long term health

In a CAFE (Clean Air For Europe) report from 2005, it is estimated that the statistical life expectancy of a European is shortened by on average 8.1 months because of exposure to the anthropogenic fraction of $PM_{2.5}$ (Amann et al., 2005). In Europe, the largest loss in life expectancy is in Belgium, with an average loss of 13.2 months. A similar study in the United States showed that a decrease in PM_{10} concentrations of 10 µg/m³ was associated with an increase in life expectancy of 0.61 ± 0.20 years (Pope et al., 2009).

The inhaled amount of particulate matter caused by exposure to ambient concentrations is many times smaller than exposure of active smokers. Long term effects of smoking, which can be considered as a worst case of exposure to air pollution, are shown by a.o. Doll et al. (2004): Cigarette smoking men, born between 1900 and 1930, died approximately 10 years earlier compared to a similar population that never smoked. Dockery et al. (1993) linked exposure to ambient $PM_{2.5}$ to all-cause mortality, cardiovascular and lung-cancer mortality for the first time in the Six US Cities Study. Almost two decennia later, exposure to ambient $PM_{2.5}$ concentrations was causally linked to cardiovascular morbidity and mortality (Brook et al., 2010; Künzli et al., 2005). No threshold level could be determined: effects are measureable even with low ambient concentrations (Lepeule et al., 2012). The evidence on long term health effects caused by air pollution has grown steadily during the last ten years. The empirically estimated dose-response function for PM is not linear, as assumed earlier, but there is a steep increase at low levels of air pollution and it flattens out with elevated levels, e.g. in active smokers (Pope, 2007, 2010). It is hypothesized by Seaton et al. (1995) that ultrafine particles are the most harmful provoking alveolar inflammation.

Susceptible groups, like children, elderly, diabetics or asthma patients, are often studied to discover effects of air pollution exposure. Gauderman et al. (2007) showed that when children lived within 500m of a motorway, this had a clear effect on lung function compared to children living more than 1500m from a motorway. Gehring et al. (2010) found a relationship between traffic-related air pollution and the prevalence of asthma in 8-year olds; several other respiratory effects have been linked to exposure to traffic-related air pollution (Laumbach and Kipen, 2012).

Due to the large number of studies looking for a relationship between air pollution and health effects, is it very hard to present a concise overview of possible effects. Different studies differ in study design, methods and techniques. In spite of the large number of studies, it is still very difficult to link exposure to a certain compound (starting from the hypothesis that not all air pollutants are equally harmful) to a specific health effect. There is an important gap in knowledge on the linkages between health effects and toxicity of pollutants or the toxicity of mixtures.

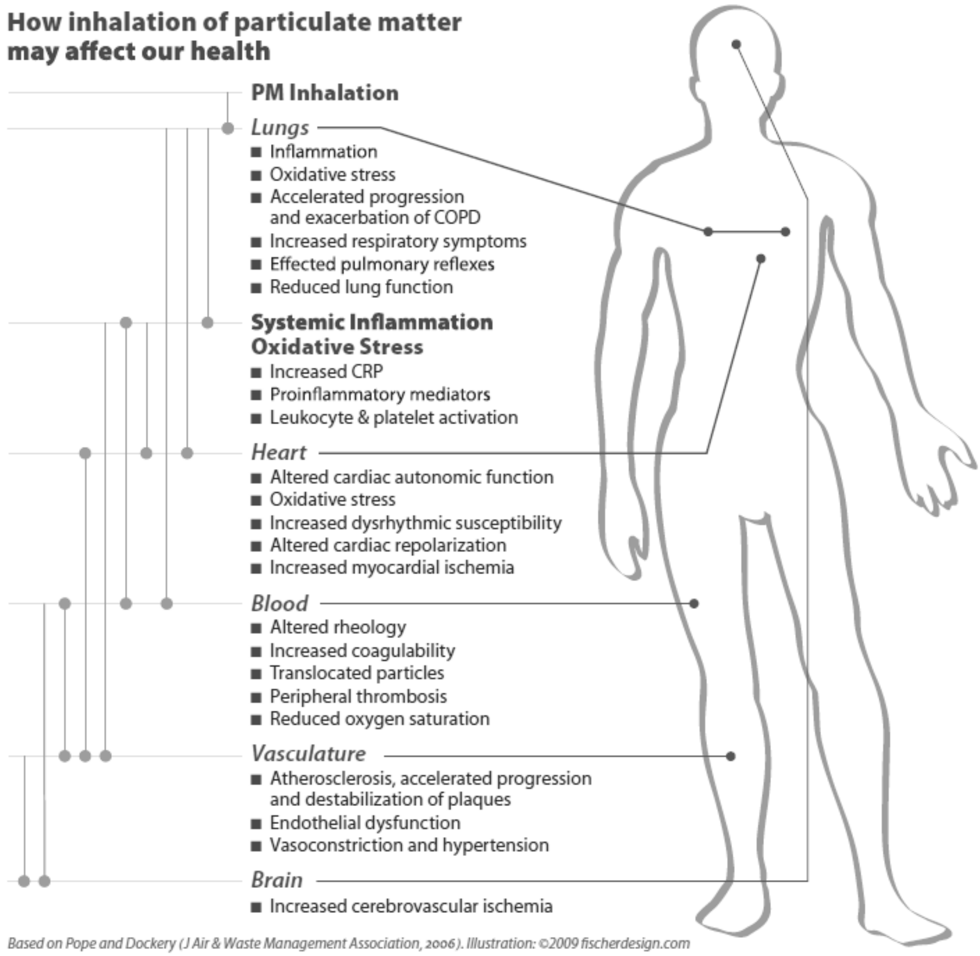


FIGURE 5: Inhalation of particulate matter and suspected health effects (Aphekom, 2011)

2.1.5 EXPOSOME

The concept of the exposome, representing lifelong exposures received by an individual from conception to death, encompasses all sources of exposure that may influence health (Betts, 2012; Rappaport, 2011; Wild, 2005). The exposome is regarded as a powerful tool to detect health effects, especially chronic diseases, caused by environmental exposures. It includes exposure via all relevant pathways: inhalation, dermal exposure, exposure through food or radiation, temperature or noise. Important confounders like stress, physical activity, or drug use are also taken into account. The exposome complements the genome: an individual gets sick either through genetic programming, through the interplay of environmental stressors, or through a combination of both.

The exposome can be determined following a bottom-up approach or a top-down approach (Rappaport, 2011). The first approach starts from environmental monitoring of air, water, food etc.: this approach measures external exposures. For air pollution exposures, this means measuring time-space paths, physical activity and air pollution sensing. The top-down approach is based upon biomonitoring of subjects, e.g. by blood sampling. It is noted by Peters et al. (2012) that when applying the top-down approach, enough attention should be paid to the linkage between the internal exposure and the external environment. Determining the exposome requires interdisciplinary research, and in the future compound-by-compound assessments should be abandoned although they may remain useful for regulation purposes (Adler et al., 2010). The exposome is an 'ideal', a 'unifying concept' rather than a true state, but it frames current research in exposure science nicely (Adler et al., 2010).

Recently, Lioy and Smith (2013) introduced the eco-exposome; it is defined as the extension of exposure science "from the point of contact between a stressor and receptor inward into the organism and outward to the general environment including the ecosphere"; thus, embracing the use of both internal and external markers of exposure.

2.1.6 CONCLUSIONS

- An ‘exposure’ has been defined as the occurrence of the event that a pollutant (at a particular concentration) comes into contact with the physical boundary of the individual, while a ‘dose’ has been defined as the occurrence of the event that the pollutant actually crosses the physical boundary (Ott, 1982).
- Concentrations measured on fixed sites are poorly correlated with true personal exposure, especially for pollutants with a high spatiotemporal variability. Inaccurate exposure assessment can lead to exposure misclassification in epidemiological studies.
- Many components are associated with some endpoint somewhere at some lag (Brunekreef, 2013). There is still an important gap in knowledge on the linkages between health effects and toxicity of pollutants or the toxicity of mixtures.

2.2 MEASURING PERSONAL EXPOSURE

Several dimensions can be identified when measuring personal exposure to air pollution. Not all of these facets need to be monitored, but the more parameters are measured, the more complete the exposure picture will look.

A first factor is **monitoring air quality**. Member states of the EU are obliged to maintain a monitoring network measuring air quality at several locations. On these sites continuous measurements take place using reference devices. When interested in personal exposures, a specific monitoring campaign should be set up with portable air quality monitors. In recent years, air quality sensors are getting a lot of attention as the ultimate means to map personal exposure to air pollution in large populations.

To enable a more thorough analysis of measured exposures, it is necessary to know which activity is executed when and where, and combine it with air quality levels. The **time-activity pattern** of an individual is unique and will differ between days; on the other hand people are subject to habitual behavior. An average daily pattern can be assumed based on large-scale time-activity studies like the Study on Travel Behavior (Onderzoek Verplaatsingsgedrag) in Flanders, NHAPS (National Human Activity Pattern Survey) in the United States, or CHAPS (Canadian Human Activity Pattern Survey) in Canada. Paper or electronic diaries can be used to get a detailed view of the diary of an individual. GPS-tracks can be collected to derive activity patterns in a relatively straightforward way.

Physical activity is important to consider when calculating the amount of inhaled air and particles. Measuring breathing rates is in most cases not realistic because of the burden of the monitors. A heart rate monitor could be used alternatively as a proxy for minute ventilation. New health sensors try to use less invasive and more objective ways to measure level of effort (Butte et al., 2012; Intille et al., 2012). Questionnaires are also being used to determine physical activity qualitatively.

2.2.1 AIR QUALITY

Air quality monitors on fixed locations are used to measure ambient air quality in a region and to check compliance with (inter)national air quality standards; by extension these monitors are used to determine exposure of a population. In Flanders the Flemish Environment Agency continuously monitors the air quality using different networks (VMM, 2010). The telemetrical network watches the overall air quality and measures the most important gaseous and particulate pollutants: ozone (O_3), sulfur dioxide (SO_2), nitrogen oxides (NO_x), particulate matter (PM_{10} and $PM_{2.5}$), carbon monoxide (CO), BTEX (benzene, toluene, ethylbenzene and xylene), and black carbon (BC). Not every pollutant is being measured at each of the 60-70 monitoring sites. Monitoring stations are spread over Flanders, and they are grouped according to location type: rural stations, suburban stations, urban stations, industrial stations and traffic stations. Most of the monitors can measure with a time-resolution of up to 1-second, but currently measurements are saved on a 30-min time base, and are soon aggregated to hourly values. The spatial resolution of these monitors is however rather low, even when considering that Flanders has the most dense monitoring network of Europe. Through spatial interpolation, an area-wide map can be produced, although for certain pollutants the accuracy of such an approach can be questioned.

Large static monitors can be used to temporarily measure air pollution in different indoor and outdoor microenvironments, rather than measuring ambient concentrations on a limited number of fixed locations (Brown et al., 2012). Several dedicated measurement campaigns already monitored concentrations in different microenvironments, but they still do not give a complete picture of personal exposure as it is impossible to measure simultaneously in all microenvironments visited by a person.

Portable devices to measure air quality exist for almost 40 years in the form of passive diffusive samplers (Yu et al., 2008). These dosimeters are still in use today to measure a.o. NO_2 concentrations; they are easy to deploy because they are small, unobtrusive, cheap and do not require power (Yu et al., 2008). Using passive samplers for monitoring results in measurements with a high spatial resolution, but due to the diffusion principle temporal resolution is low (typically a few days to a few weeks). Different types of passive samplers exist, with Palmes tubes, Yanagisawa filter badges, Ogawa samplers and Radiello samplers being the most famous (Yu et al., 2008).

Knowing where and when exposure occurs is crucial to determine causality between air pollution sources and health effects (Adams et al., 2009; HEI, 2010). Active pumped samplers can measure with a much higher temporal resolution than passive samplers, and they can therefore detect air pollution peaks. Active monitors are expensive, require expert handling, and are often too large to use in true personal monitoring. Examples of popular portable active monitors include the DUSTTRAK aerosol monitor (PM_{10}, $PM_{2.5}$, PM_1), GRIMM aerosol monitor (PM_{10}, $PM_{2.5}$, PM_1), P-TRAK ultrafine particle counter (UFP), aethalometer (BC), Aerasense ultrafine nanoparticle monitor (UFP), particle soot absorption photometer PSAP (soot), and personal DataRAM (PM_{10}). To limit possible confounding, the inlet of the monitor can be placed near the breathing zone of the individual (Adams et al., 2009; Broich et al., 2012; Fruin et al., 2004).

The ultimate air quality monitor can measure with a high temporal and spatial resolution, is affordable and is as little obtrusive as possible. A high spatial resolution can be achieved through either the portability of the instrument, or because monitors can be spread in large quantities across a study area. Air quality sensors can fill this gap (Mead et al., 2013). They measure with a high temporal resolution, sensors are small and portable, require no or little power, are cheap and quiet. Sensors that are commercially available for the moment are not yet reliable enough for portable deployment; it is especially very difficult to reproduce measurements from a reference monitor. Sensors used to be developed to measure peak concentrations, e.g. in vehicle exhaust, but they are not sensitive enough for detecting ambient concentration levels. Sensors that proved to be relatively good, e.g. CO-sensors, are unfortunately not very relevant for health at current ambient concentrations.

By deploying air quality sensors in large quantities, a wealth of data could be collected. Measurements with sensors should be combined in a database, and visualized, e.g. using Google Earth. Sensors communicate with a server and with each other; these systems are referred to as air quality sensor networks (Cordova-Lopez et al., 2007; Richards et al., 2006). In the future, individuals will be equipped with sensors measuring personal exposure to different air pollutants, and they will be able to consult and visualize the results themselves. Peak exposures can be detected or sources of air pollution could be identified: this information is valuable for an individual, but it could also help governments and scientists (Lioy and Smith, 2013). Empowering people to do research in their own environment, or 'participatory sensing' as it is called, has huge potential and is identified as a major research point in achieving the exposure science vision for the 21st century (Conrad and Hilchey, 2011; Lioy and Smith, 2013; Loh et al., 2002).

2.2.2 TIME-ACTIVITY PATTERNS

One of the most important theories on the movement of people in time and space is the theory of Torsten Hägerstrand (1970). The time-geography theory describes consecutive activities on different locations, and the trajectories that link this activity chain. The space-time path can be represented graphically in a three-dimensional space, the 'space-time aquarium' (Kwan and Lee, 2003). A 24h-period is presented in the aquarium as a continuous temporal sequence in geographical space (FIGURE 6). Individuals are limited while performing their daily activities by the amount of time available: their time-budget. Other constraints in time and space, linked to mandatory activities (e.g. work) or through interaction with other individuals, describe the potential activity-space of an individual.

The emergence of geographical information systems has resulted in a materialization of the theories of Hägerstand in the '90 (Briggs, 2005). But to draw the 'lifelines' of an individual and gain insight into space-time paths, a time-activity survey needs to be conducted.

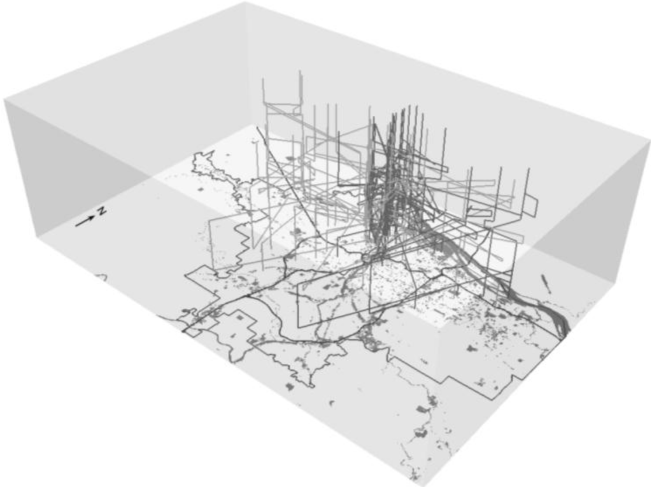


FIGURE 6: Space-time aquarium with 'lifelines' (Kwan and Lee, 2003)

Collecting time-activity data lays a heavy burden on respondents. Filling in questionnaires, either on paper or in electronic format, asks time and dedication of the participants, and thus there is a high risk of non-participation or drop-out. A high degree of participation should be aimed for, e.g. by being respondent-oriented (study design for example according to the New Kontiv Design), with clear questions, and with a follow-up of the participants. However, differential non-response does occur resulting in bias in activity pattern surveys, e.g. in lower socio-economic classes, short walking trips are not reported, busy participants with a lot of trips, children or elderly (Arentze et al., 2000).

Traditionally time-activity patterns are collected with paper and pencil surveys (a.o. Dadvand et al. (2012) in an epidemiological study), sometimes supplemented with a telephone interview (a.o. Axhausen et al. (2002) and Delgado-Saborit (2012)). A reward or incentive can be presented to participants at the end of the survey. A major disadvantage of paper diaries is the poor data quality: gaps, overlaps and missing values frequently emerge in the diaries; the quality can be enhanced by going across the diary with the participant. Additional errors can be introduced when manually entering the data in a database. An advantage of a paper-and-pencil diary is that it can be filled in at any time and at any place. Paper diaries can be aided by a voice recorder handed to the participants (Delgado-Saborit, 2012).

Electronic diaries can be imputed in a computer directly by participants. iCHASE (Internet Computerized Household Activity Scheduling Elicitor (Lee et al., 2000)) was developed to enhance data quality (e.g. built-in consistency checks) and improve user guidance. A web-survey is cost-efficient and can reach a lot of participants in a short time span (Wu et al., 2011b). Diary data is immediately available; manual data-input is subject to human error and can be avoided when using electronic diaries. Mobile electronic diaries combine the advantages of the paper and pencil method, namely portability, with the advantages of an electronic survey. A first application of software to register activities and trips on a palm-sized computer or PDA was 'EX-ACT' (Rindsfüser et al., 2003), based on the previous iCHASE desktop software. Kochan et al. (2010) developed custom-designed software 'PARROTS' to register activities on a PDA. Delfino et al. (2010) also used a PDA and every hour participants had to indicate which activity they were performing; the questionnaire was supplemented with questions on the emotional state and the level of physical activity. In the two latter studies, paper diaries were used instead of electronic diaries if preferred by the respondents.

A GPS logger can be used complementary with a diary or as a means in itself. Gerharz et al. (2009) and de Nazelle et al. (2013) combined GPS loggers with paper diaries; Kochan et al. (2010) enriched data from an electronic diary with a built-in GPS receiver. Bohte and Maat (2009) and Wu et al. (2011a) solely used GPS tracks to trace back activity patterns and transport modes by using data fusion approaches. A web-interface was built to enable participants to validate the derived activities and trips, similar to Beusen et al. (2009) and Beckx et al. (2013). A GPS logger can track exact routes, although the GPS signal can be distorted or lost in urban and indoor environments (Beekhuizen et al., 2013; Wu et al., 2010).

Each of the methods to collect time-activity diaries has its pros and cons. Biases or errors, like human error, recall error or other cognitive challenges, social desirabilities, loss of GPS signal, are a larger problem in some of the methods. The longer the duration of the survey, a fatigue effect can be introduced: less activities or trips will be registered than performed in reality; in a worst case, participants will drop out. Usually a time-activity survey lasts for 1 day up to 1 week, sometimes repeated in a contrasting season.

2.2.3 PHYSICAL ACTIVITY

Level of physical activity can be examined through self-reports or through wearable sensors. Several surveys exist that estimate physical activity by means of paper questionnaires (Baecke et al., 1982; IPAQ, 2005). Sensors can be grouped in two categories: monitors that register movement, and monitors that register body functions.

Movement can be detected with GPS, accelerometers, or step counters. Accelerometric sensors can be placed on the body and they record 3 axes of movement (Rodes et al., 2012). Accelerometers are less sensitive for activities that incorporate increased skeletal movement, such as walking uphill (Delfino et al., 2010; Pober et al., 2006). More importantly, poorly conditioned subjects may experience a higher level of perceived exertion because relatively low levels of physical activity measured by the accelerometer are closer to their maximal level of performance than for subjects who are physically fit (Delfino et al., 2010). As an alternative of placing sensors on the body, accelerometric sensors already present in a smartphone can be used (de Nazelle et al., 2013).
Monitors that register body functions include a wide range of devices, from heart rate monitors, over face masks to novel integrated health sensors. Zuurbier et al. (2011) used heart rate monitors to measure level of effort while cycling; Holter monitors were used by Weichenthal et al. (2011) to continuously record electrocardiograms. Int Panis et al. (2010) measured minute ventilation, breathing frequency and tidal volume using a portable cardiopulmonary indirect breath-by-breath calorimetry system fixed into a chest harness. An actigraph placed near the waist combines movement sensors (accelerometer) and health sensors (heart rate) (Delfino et al., 2010; Patterson et al., 1993; Rodes et al.,

2012). Additional physiological signals such as skin temperature, galvanic skin response, respiration rate, and foot pressure might be useful for improving physical activity or energy expenditure estimation when used with accelerometery or heart rate monitoring, and for monitoring wear time (Arvidsson et al., 2007; Butte et al., 2012; Intille et al., 2012). Several methods exist to objectively estimate ventilation or inhalation rate using activity monitors that can easily be deployed in the field (Dinesh et al., 2013).

2.2.4 CONCLUSIONS

- Because human activities are directly related to the timing, the location and the degree of pollutant exposure, all those factors have an important role in explaining variations in exposure (Klepeis et al., 2001).
- Air pollution sensors are promising, but comparability of the measured pollution levels between sensors and reference monitors is still low.
- Electronic diaries are being used to register time-activity patterns, but more traditional paper and pencil diaries are still used interchangeably.
- A wide variety of physiological sensors, including monitors that measure movement or body functions, are commercially available to estimate physical activity and ventilation.

2.3 MODELING PERSONAL EXPOSURE

The most straightforward method to model personal exposure to air pollution is combining a concentration map with high spatial resolution with the residential location of an individual. But this is only part of the story: individuals as well as pollutants are in constant motion. Therefore, personal exposure to air pollution should be modeled as a combination of two interacting geographies: a moving population and a continuously changing air quality (Briggs, 2005).

By using geographical information systems, it is possible to develop exposure models that take into account population movement and changing pollution fields. **Time-activity patterns** and movement of people were formalized by Hägerstrand (1970): he developed the concept of individuals moving through space and time. Modeling time-activity patterns starts with the registration of hundreds of diaries, followed by the extrapolation of these diaries to the full study population. Activity-based traffic models estimate whereabouts and trips for a synthetic population and will be discussed in more detail in this chapter.

Concentration fields with a high spatial resolution can be produced using different techniques: geostatistical interpolation based on measurements on fixed sites, dispersion modeling, land use regression, or hybrid modeling techniques (HEI, 2010; Jerrett et al., 2005a). Modeling concentrations with a high temporal resolution is often not aimed at, but hourly or daily measurements on fixed locations could be used, and also several examples exist of hourly dispersion models. Land use regression models will be applied in this PhD and current state-of-the-science is therefore presented hereafter.

Existing **modeling frameworks** estimating population and/or personal exposure are reviewed in a third paragraph. These models have in common that the exposure is defined as a function of time spent in location *i* combined with the concentration on that location. But datasets used are diverse; some models need input from revealed diaries or GPS-tracks, while others are completely model-driven.

2.3.1 ACTIVITY-BASED MODELS

Traditional transport models are trip-based: trips are considered as isolated events; the reason why people are traveling is ignored. Relationships between different trips are non-existent. Four aspects of travel are predicted separately: the number of trips, the origin and the destination of each trip, the transport mode, and the route. These so-called 4-step models have important limitations: through the spatial, demographic, and temporal aggregation it is impossible to analyze trip patterns on a disaggregated individual level (Kitamura et al., 2000). The sequential model structure results in little dynamics, and policy scenarios produce unrealistic outcomes.

Because of the obvious limitations of 4-step models, tour-based models and finally activity-based models emerged (Axhausen and Gärling, 1992; Davidson et al., 2007; Kitamura et al., 2000). The main strength of these models is that the demand for trips is derived from activities that individuals wish or need to perform. A point of difference with trip-based models is that sequences of trips and activities are modeled: first activity x is performed, then your car takes you to the next activity while en route you stop at a shop, finally you return home using the same car. Spatial, temporal, personal and household, institutional, and transport constraints are taken into account in the modeling. This results in a model that estimates which activities are conducted where, when, for how long, with whom, the transport mode involved, and preferably also the route.

Activity-based models are better suited for an evaluation of transportation control measures than 4-step models because secondary effects can be included in the analysis (Shiftan, 2000). Secondary effects are adjustments to the activity pattern because of a primary effect. For example, cheaper public transport can convince a commuter to switch from car to train: this is the primary effect of a public transport subsidy. But the commuter may have to make an additional trip for shopping, if it is impossible to do this on his way home: this is the secondary effect.

Most activity-based models start modeling activities at 3 a.m. until 3 a.m. the next day. This approach was introduced by Kitamura et al. (2000) as 98% of all participants filling out a diary indicated that they were at home at 3 a.m. Activity-based models are sensitive for time-of-day: all activity-based models have used at least 4 network assignment periods; but in most cases more precise time windows are used, e.g. 60-minute time intervals (Bradley and Bowman, 2006). Diaries are predicted for all days of the week; seasonal effects are included in only a minority of the activity-based models (Henson and Goulias, 2006; Kitamura et al., 2000).
Activity-based models spatially aggregate data to the level of 'zones'. Origin/destination matrices are made for TAZs or Traffic Analysis Zones (an individual travels from zone A to zone B, or performs an activity in a zone). Data is not available on a finer scale to more realistically model activity patterns. For example, from the national statistics agency data are available that describe how many people work in a certain zone; but data cannot be retrieved on which individual living in a specific house works in that specific zone.

The characteristics of an activity-based model are summarized by McNally (2000):

1. Travel is derived from the demand for activity participation;
2. Sequences or patterns of behavior, and not individual trips, are the relevant unit of analysis;
3. Household and other social structures influence travel and activity behavior;
4. Spatial, temporal, transportation, and interpersonal interdependencies constrain activity/travel behavior;
5. Activity-based approaches reflect the scheduling of activities in time and space.

Several activity-based models have been developed: STARCHILD (Axhausen and Gärling, 1992), Portland (Shiftan and Suhrbier, 2002), AMOS/FAMOS (Pendyala et al., 2005), TASHA (Miller and Roorda, 2003; Roorda et al., 2007), ALBATROSS (Arentze and Timmermans, 2004). Henson and Goulias (Henson and Goulias, 2006) and McNally (2000) give an overview of existing activity-based models and their characteristics.

For Flanders, FEATHERS (‘Forecasting Evolutionary Activity Travel of Households and their Environmental RepercussionS’) was developed (Bellemans et al., 2010). FEATHERS is a simulation platform for implementing activity-based models in a predefined geographical area. FEATHERS has a module-based design, is agent-based and object oriented. By focusing on the disaggregated level of an individual, the added value compared to 4-step models is clear. Each member of the population is represented in the FEATHERS platform as an agent. Comparable to a real-life person, an agent autonomously performs certain activities during a simulation.

2.3.2 LAND USE REGRESSION MODELS

In a land use regression (LUR) model statistical associations are developed between potential predictor variables and measured pollutant concentrations as a basis for predicting concentrations at unsampled sites (Hoek et al., 2008; Ryan and LeMasters, 2007). Originally, this technique was called regression-mapping and it was first applied for NO_2 to estimate the long-term exposure in the SAVIAH project in four urban areas (Briggs et al., 1997).

Air quality monitoring should take place on 20-100 different sites (Basagaña et al., 2012; Hoek et al., 2008). Jerrett et al. (2009) used measurements on 143 locations to form a regression model for the city of Toronto. Occasionally routine air quality networks are used for this purpose, but because of the limited spatial density of these networks, a purpose-designed measurement campaign is often set up. Ryan and LeMasters (2007) showed that the fit (R^2) of the final model not only depends on the number of measurement sites, but also on the positioning; this is confirmed by Johnson et al. (2010). There is no formal methodology to determine the minimum number of measurements required to build a proper land use regression model (Poplawski et al., 2009). When a monitoring campaign is designed, measurement sites should be as diverse as possible with respect to expected concentration levels (Lebret et al., 2000). Moreover, measurement sites should be representative for the targeted study population (Wang et al., 2012). The selection of sites can be done using the formal location-allocation method (Kanaroglou et al., 2005), but in many cases an expert opinion is considered to be sufficient. Sites are often grouped according to location type, e.g. regional background sites (no impact of local sources within several km²), urban background sites (no impact of local urban sources, more than 50m away from major roads), traffic sites (located near a major road). If measurements on the different sites cannot run simultaneously, a correction for changing background concentrations should be made (Brauer et al., 2006). If there is not enough spatial variability in the sites, the final LUR model will not perform well on a local scale. Hystad et al. (2011) tried to compensate for that by adding deterministic gradients to capture local-scale variation based on the proximity of certain sources (i.e. traffic, industrial sources).

Each measurement site is geocoded and for each coordinate a buffer with radius x is considered. Other forms than a circular buffer have been used, e.g. a wig (taking into account wind speed and wind direction) or a square (moving window technique (Vienneau et al., 2009)). The size of the buffer radii should depend on the pollutant and on the variable, but it is generally within 20m up to 10km. Buffer sizes should be based on the decay rate of a specific pollutant away from sources, e.g. buffers for $PM_{2.5}$ should be wider than buffers for diesel particles; as well as on the density of the geographic variables surrounding the sampling location (Ryan and LeMasters, 2007; Su et al., 2009). If several buffers of the same variable are selected for inclusion in the final regression function, it is possible to keep only the inner buffer and use concentric rings or donuts for the wider buffers. The use of nested buffers or the donut-model will not affect the form of the final LUR model (von Klot, 2011).

Independent variables that can be used in LUR models are traffic variables, land use variables, population variables, meteorology and geography, and possibly other variables (street canyon, season, distance to point source, concentration of other pollutants or noise maps). Neither of the variables needs to be forced in the model, but not including traffic variables when modeling a traffic-related air pollutant will result in a LUR model with little predictive power.

A widely used algorithm to build a land use regression model is presented in Henderson et al. (2007).

(1) Rank all variables by the absolute strength of their correlation with the measured pollutant.

(2) Identify the highest-ranking variable in each sub-category.

(3) Eliminate other variables in each sub-category that are correlated (Pearson's r >= 0.6) with the most highly ranked variable.

(4) Enter all remaining variables into a stepwise linear regression.

(5) Remove from the available pool any variables that have (a) insignificant t-statistics (α=0.05) and/or (b) coefficients that are inconsistent with a priori assumptions.

(6) Repeat steps 4 and 5 to convergence and remove any variable that contributes less than 1% to the R^2 value for a parsimonious final model.

Adaptations to this methodology are possible, e.g. Johnson et al. (2010) initially removed variables that were highly correlated with another variable to prevent multicollinearity in regression models.

The application of the methodology presented above, or alternative methods, results in a multiple linear regression model of the form:

$$Concentrations\ pollutant = \alpha + \beta_1 X_1 + \beta_2 X_2 + \beta_3 X_3 - \beta_4 X_4$$

α is the model intercept; β_1 is the coefficient of X_1, for example a variable for population density in a buffer with radius 500m; β_2 is the coefficient of X_2, e.g. a variable for traffic intensity on the nearest road; β_3 is the coefficient of X_3, e.g. a variable for industrial land use within 5km; and β_4 is the coefficient of X_4, this can be for example altitude. A LUR model has on average 3 to 6 independent variables.

Air pollution models can be validated using different methods. LUR models are most often validated using leave-one-out cross-validation: the regression model is calibrated using n-1 locations and predicted concentrations are compared to observed concentrations on the n^{th} location. This procedure is repeated n times and gives an indication of the performance of the model (Beelen et al., 2013; Brauer et al., 2003; Eeftens et al., 2012; Hoek et al., 2008; Johnson et al., 2010). The leave-one-out cross-validation procedure should be used with caution if the number of sites is small (25 or less): cross-validation overestimates the predictive power of the model through overfitting (Wang et al., 2012). If enough measurement sites are used, hold-out validation is a good option. Observations not used for model fitting, are compared to concentrations predicted by the LUR model (Briggs et al., 1997; Johnson et al., 2010; Mölter et al., 2010a; Ryan et al., 2008). Two strategies are possible: part of the measurements from the campaign is left-out, or an external dataset is used (Wang et al., 2012). As a third option for LUR model validation, concentrations measured at the official air quality network can be compared to predictions of the LUR model (Brauer et al., 2006; Henderson et al., 2007; Mölter et al., 2010a). Statistical indicators to evaluate air quality models include root mean square error (RMSE), bias, standard deviation (SD) and the correlation coefficient (r) (Thunis et al., 2012).

Once the regression model is validated, the model can calculate a concentration for every point in the study area. In epidemiological studies, a concentration will be estimated for the residential location of each member of a cohort. Concentrations can also be calculated for additional locations where participants spend a considerable amount of time (Ryan et al., 2008). A concentration map can be produced by calculating concentrations for centroids of grid cells in a fine raster.

Most LUR models estimate annual concentrations. Only a handful of models try to include temporal variation; e.g. calculating concentrations for previous years or through the inclusion of daily concentrations. Nethery et al. (2008a) and Slama et al. (2007) developed one annual LUR model and applied a correction factor that is representative for differences between seasons. Crouse et al.

(2009) made three different LUR models (winter, spring, summer) by doing measurements in different seasons and calculating separate models. From the previous studies, it appeared that concentration levels changed between periods, but the geographical pattern was very similar (hot spots versus cold spots). Most LUR models are built for an urban area; only a few models considered a national or transnational scale (Beelen et al., 2007; Beelen et al., 2009; Janssen et al., 2008; Stedman et al., 1997; Vienneau et al., 2010).

Alternatives to LUR models are dispersion models, geostatistical interpolation based on measurements on fixed sites, or hybrid models. Several studies compared LUR models with dispersion models and the performance of these models is similar (Beelen et al., 2010; Briggs, 2007; Briggs et al., 2000; Clougherty et al., 2008; Cyrys et al., 2005; Dijkema et al., 2011; Hoek et al., 2008; Liu et al., 2012). Mercer et al. (2011) compared universal kriging with LUR: the R^2 and RMSE perform slightly better in the universal kriging, but the performance of the LUR model is also acceptable. Gulliver et al. (2011a) examined the performance of 10 different methods to assess exposure to PM_{10}. Simple models (distance to the nearest road, traffic intensity on this road, fraction of heavy traffic, road density and traffic intensity within buffers, etc.) and more complex models (nearest fixed monitor, kriging, dispersion modeling, land use regression modeling) were developed, but only the LUR technique could predict concentrations on a fixed location with enough reliability. Simple techniques like interpolation or inverse distance weighting are based on Tobler's First Law of Geography: “Everything is related to everything else, but near things are more related than distant things”. This does not always hold true in air pollution mapping: e.g. with strong winds concentrations will be higher on one side of the street, or in the case of chimneys concentrations on ground level will not be elevated until several 100m further (Briggs, 2005).

2.3.3 EXPOSURE MODELS

SHEDS / EMI SHEDS or the Stochastic Human Exposure and Dose Simulation Model (Burke et al., 2001) estimates average personal exposure based on ambient concentrations measured at a fixed monitoring station. Concentrations differ between different microenvironments because of changing air exchange rates. Activity diaries are composed for the study population and depend on personal characteristics. Diaries originate from the Consolidated Human Activity Database (CHAD) from U.S. EPA. Persons with the same characteristics will have the same agenda which is a drawback of this approach. Personal exposure is then modeled by multiplying the time spent in a microenvironment with the concentration in that type of microenvironment. The added value of this approach was in (1) the use of time-activity patterns instead of using a non-mobile population, and (2) the use of air exchange rates.

Recently EMI (Exposure Model for Individuals) was developed by the U.S. EPA in succession to SHEDS (FIGURE 7) (Breen et al., 2010). The approach is very similar, but both models differ in aim / applications. SHEDS primarily focuses on the exposure of a population using time-activity patterns from CHAD, whereas EMI is based on GPS tracks and diaries of individuals. The algorithms to calculate inhaled dose and indoor air quality are similar to the submodels used in SHEDS.

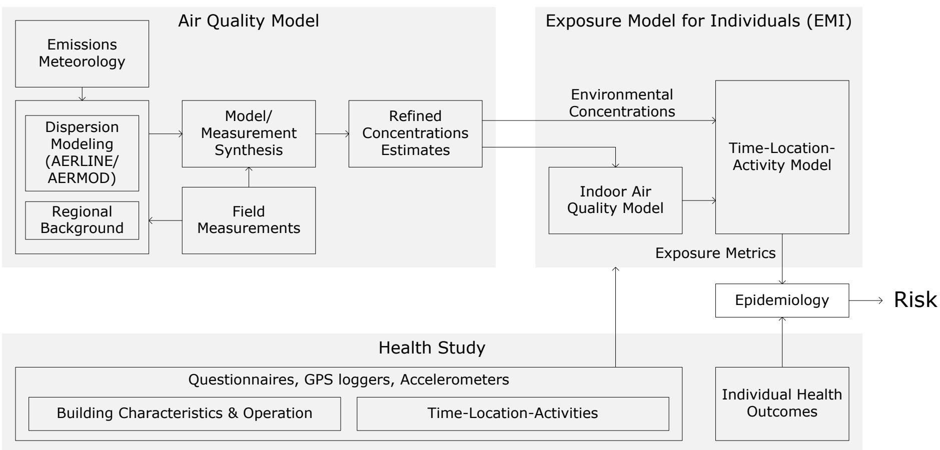


FIGURE 7: EMI – Exposure Model for Individuals (Vette et al., 2013)

pCNEM pCNEM (Probabilistic version of the Canadian version of NAAQS Exposure Model) or its earlier versions NEM and pNEM aim at predicting personal exposure (Zidek et al., 2003; Zidek et al., 2005). Personal characteristics (e.g. socio-economic data), ambient pollution levels, and the activity pattern of individuals all form the input of the pCNEM. Per population subgroup the same time-activity pattern is generated, originating from the National Human Activity Pattern Study (NHAPS). A major difference with SHEDS is the calculation of concentrations in different microenvironments: an individual is assigned to two grid cells; the first grid cell contains his residential location and the second grid cell his work location. The geographic zones are relatively large leading to unrealistic predictions on a fine spatial scale. Transport and exposure in different microenvironments are not taken into account.

STEMS STEMS (Space-Time Exposure Modeling System) is a GIS-based dynamic exposure model for particles that estimates personal exposure on locations and while traveling (Gulliver and Briggs, 2005; Gulliver and Briggs, 2011). STEMS includes four submodels: the traffic model SATURN, a dispersion model ADMS, a model estimating background concentrations, and a time-activity-based exposure model TOTEM (FIGURE 8). From the traffic model emissions are calculated and used as an input in the dispersion model; afterwards hourly pollution maps are derived. 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. The exposure model TOTEM takes into account the pollutant concentration during time *i* at location *j*, multiplied with a weighting factor specific for different environments. Exposure while traveling on foot or by bike is determined by the ambient concentration at that location. For in-vehicle exposure, a linear relationship between ambient and in-vehicle concentrations is assumed.

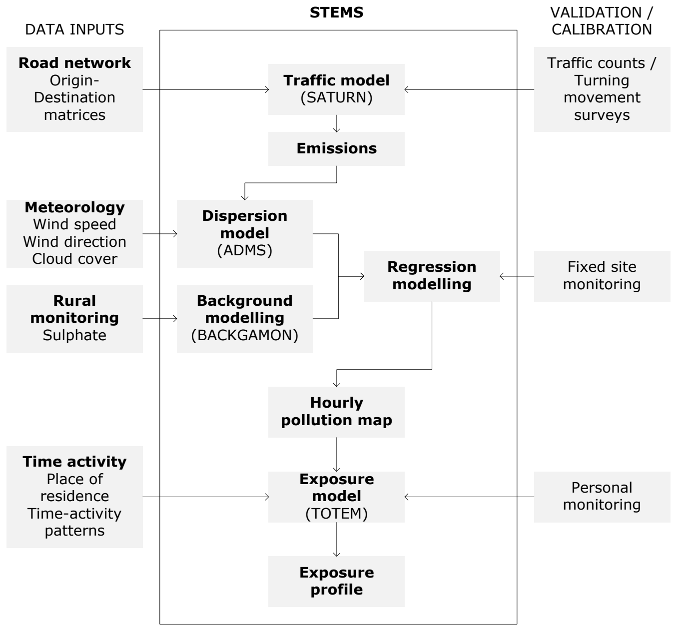


FIGURE 8: STEMS - Modeling structure (Gulliver and Briggs, 2005)

SESM (Setton et al., 2008) Personal exposure is determined with SESM (Spatial Exposure Simulation Model) on the basis of concentrations and time spent in six microenvironments (home indoor, work indoor, other indoor, outdoor, in vehicle to work, in vehicle other). The time spent in each environment is derived from CHAPS (Canadian Human Activity Pattern Survey), and includes a stochastic component to be able to produce different diaries for people with the same characteristics. The activity data is then combined with geographical info on the location of work places in the study area. Individual trips are assigned to a road network according to the shortest path algorithm. Intrazonal trips and trips with origin or destination outside of the study area are excluded. An existing LUR model for NO_2 was applied to estimate concentrations for microenvironments in each geographical zone. If individuals were in transport, concentrations in subzones were time-weighted for trips crossing different subzones. Indoor concentrations are determined by applying indoor/outdoor ratios from previous studies, and are specific for each microenvironment.

BESSTE (de Nazelle and Rodríguez, 2009; de Nazelle et al., 2009) The BESSTE (Built Environment Stochastic Spatial Temporal Exposure) model simulates energy expenditure and inhaled dose of air pollutants (PM10 and ozone) for residents of Orange County, North Carolina. The inhaled dose for the exposure event is defined as the product of the associated ventilation rate, the concentration of the pollutant in the microenvironment in which the activity takes place, and the duration of the event.

The modeling framework first develops diaries for each individual; activity patterns are drawn from the Consolidated Human Activity Database (CHAD) from U.S. EPA (FIGURE 9). Secondly, for every activity geographic locations are chosen using gravity models. A transport choice model is applied to predict the transport mode for a particular trip. Air pollutant concentrations are modeled by combining different sources of space-time information, including monitoring data, photochemical model outputs of ambient air pollution, and models of micro-scale dispersion around traffic roads.

One of the limitations of the BESSTE model is that the air pollution map is 'external', meaning that changes in the activity pattern (e.g. more pedestrian friendly neighborhoods) do not change air quality. Like most other exposure models, the BESSTE model is not yet validated using personal measurements.

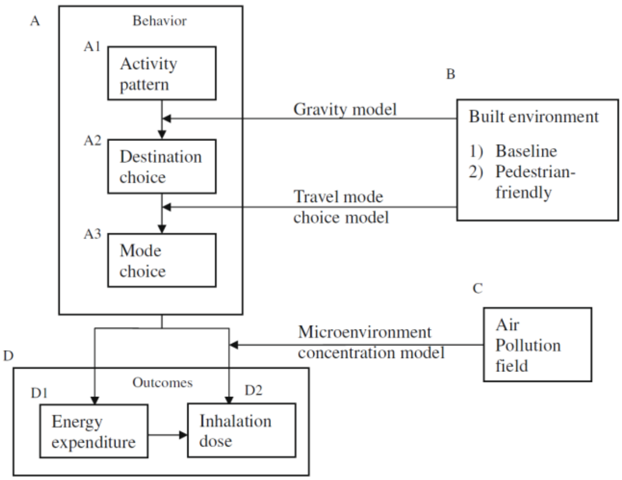


FIGURE 9: BESSTE - Built Environment Stochastic Spatial Temporal Exposure model (de Nazelle and Rodríguez, 2009)

Activity-based model / dispersion model In this study an activity-based model was used to estimate population exposure to NO_2 (Beckx et al., 2009a; Beckx et al., 2009b; Beckx et al., 2009c). Besides the activity-based model for the Netherlands ALBATROSS, an emission model (MIMOSA) and a dispersion model (AURORA) were used (FIGURE 10). On the basis of traffic streams, which are an outcome of the activity-based model, spatially dispersed emissions were calculated. Hourly emissions served as an input for the dispersion model. Personhours in each zone from ALBATROSS were combined with hourly concentrations, and 'dynamic' population exposure was estimated. Policy scenarios can be calculated using this framework: changes in the activity pattern (e.g. because of fuel price increase, changing shop opening hours, etc. (Dhondt et al., 2012b; Dons et al., 2011a)) will impact both individual diaries, concentrations and exposure. A very similar approach was followed by Hatzopoulou et al. (2011) for the Greater Toronto Area.

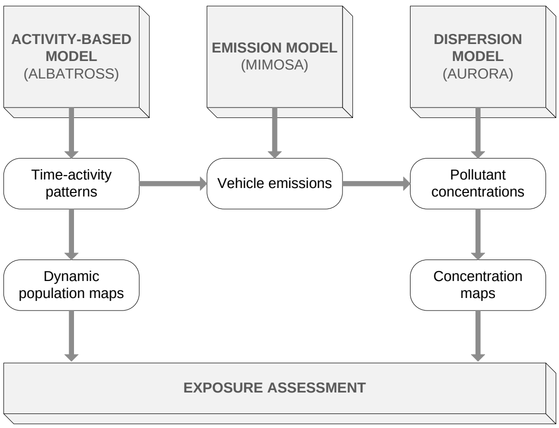


FIGURE 10: Schematic overview of the integrated activity-based modeling framework (adapted from Beckx et al. (2009a))

MEEM MEEM (MicroEnvironmental Exposure Model) aims to simulate exposure to NO_2 by combining time-activity patterns, modeled outdoor and indoor concentrations (FIGURE 11) (Mölter et al., 2012). Outdoor concentrations are modeled with a LUR model; temporal variation is added by applying the hourly concentration pattern of a background monitor. The INDAIR model was parameterized for the UK and applied to estimate indoor concentrations in multiple microenvironments. Exposure in transport was estimated using the same LUR model: each road segment was cut in 100 segments, on the center point of each of these segments the LUR model was run, finally the shortest route between two locations was calculated and was intersected with this polyline map of pollution (Mölter, 2012, personal communication). MEEM is validated by measuring personal exposure of 60 children to NO_2 with passive samplers while registering the time-activity pattern in paper diaries. MEEM could predict personal exposure more accurately than ambient concentrations measured on a fixed monitoring location, and better than modeled LUR concentrations at the home location of the child.

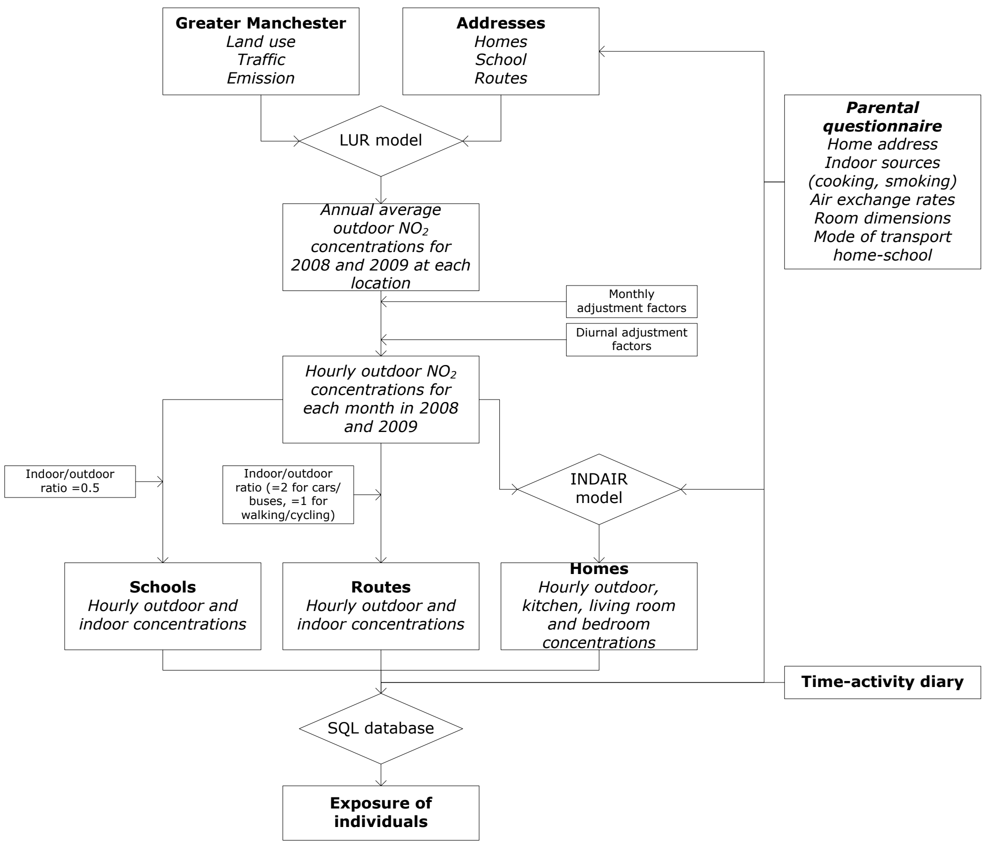


FIGURE 11: MEEM – Flow diagram showing input data, modeling steps, intermediate and final output datasets (Mölter et al., 2012)

Personal exposure model Münster Personal exposure to air pollution is modeled using personal activity profiles, ambient air quality, and an indoor model (FIGURE 12) (Gerharz et al., 2013). Time-activity diaries were collected using paper diaries (to detect exposure-relevant activities with indoor emission sources), and GPS-loggers (to provide geographical positions) for 10 individuals. Ambient PM_{10} concentrations were modeled using a combination of kriging interpolation and a Lagrangian air pollution dispersion model. Indoor concentrations are estimated with a mass-balance model; exact parameter values for e.g. air exchange rate, room volume, and deposition rate cannot be achieved, so parameter distributions were derived from available studies in other countries. For the transportation environment, the distribution of the ratio between in-vehicle and ambient concentrations as measured in different exposure studies is multiplied with the ambient concentration modeled in Münster. The model approach is deployed as a web service to enhance accessibility and reusability.

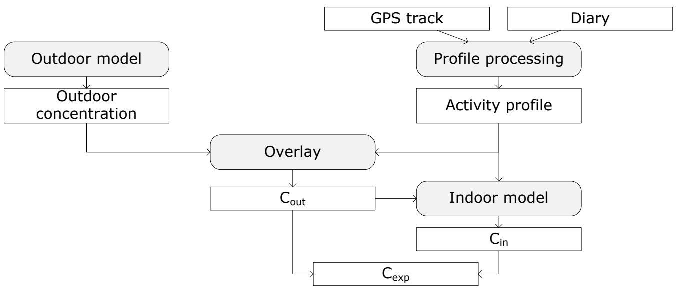


FIGURE 12: Exposure modeling methodology as applied in Münster, Germany (Gerharz et al., 2013)

2.3.4 CONCLUSIONS

- Many techniques exist to capture temporal and spatial variability in air pollution concentrations (interpolation, land use regression, dispersion modeling). Simulating time-activity patterns is more difficult; activity-based models are suitable but the spatial aggregation in zones contrasts with the highly accurate air pollution fields.
- Activity-based models are not yet widespread; many areas do not have a working activity-based model. Developing such a model requires expert knowledge, is data-intensive, and cannot realistically be done in an exposure or epidemiological study.
- Land use regression models become increasingly popular due to the many advantages of the model. Moreover, it appeared that land use regression models perform as good as or better than other more established techniques as kriging or dispersion modeling.
- Exposure models such as SHEDS or pCNEM that estimate exposure, are valuable, especially when compared to the use of concentrations measured with fixed air quality monitors. Still many of these models make unrealistic assumptions.
- SHEDS and pCNEM are examples of modeling frameworks that estimate population exposure. STEMS, BESSTE, MEEM and EMI are examples of models that estimate personal exposure. Most models estimating exposure of individuals are up till now not designed to model exposure of a full population as specific information on individuals, not available on an aggregated level, is required.
- An important issue is that the final outcome of population exposure models has never been fully validated; the validation was limited to the validation of submodels. For population exposure models, this means that the space-time predictions need to be validated as well, together with the air quality models.

2.4 BLACK CARBON

Soot, elemental carbon (EC), black smoke (BS), refractive carbon, light absorbing carbonaceous PM, absorption coefficient, diesel particulate matter (DPM): all these terms are used synonymously for black carbon (BC). But in fact they should not be used interchangeably as they are all measured and defined differently. Universally accepted nomenclature is still lacking; in recent documents, BC is defined as:

- BC is often used interchangeably with soot, but more recently has been operationally defined as those PM emissions that are quantified in the exhaust using light attenuation techniques (CARB, 2010).
- Operationally defined aerosol species based on measurement of light absorption and chemical reactivity and/or thermal stability (UNEP, 2011).
- BC can be defined specifically as a solid form of mostly pure carbon that absorbs solar radiation (light) at all wavelengths (U.S.EPA, 2012b). BC is the most effective form of PM, by mass, at absorbing solar energy. BC is a major component of "soot", a complex light-absorbing mixture that also contains organic carbon (OC).
- BC is an operationally defined term which describes carbon as measured by light absorption (WHO, 2012).
- BC refers to the dark, light-absorbing components of aerosols that contain two forms of elemental carbon (Han et al., 2007; WHO, 2012):
    - char-EC: the original graphite-like structure of natural carbon partly preserved, brownish color;
    - soot-EC: the original structure of natural carbon not preserved, black color.
- Black carbon is a distinct type of carbonaceous material, formed only in flames during combustion of carbon-based fuels (Bond et al., 2013). It is distinguishable from other forms of carbon and carbon compounds contained in atmospheric aerosol because it has a unique combination of the following physical properties:
    - It strongly absorbs visible light with a mass absorption cross section of at least 5 m^2g^{-1} at a wavelength of 550 nm.

- It is refractory; that is, it retains its basic form at very high temperatures, with a vaporization temperature near 4000K.
- It is insoluble in water, in organic solvents including methanol and acetone, and in other components of atmospheric aerosol,
- It exists as an aggregate of small carbon spherules.

2.4.1 PROPERTIES OF BLACK CARBON

Generally, more than 90% of BC is in the $PM_{2.5}$ size fraction (Viidanoja et al., 2002). BC contributes 5-10% to $PM_{2.5}$ and somewhat less to PM_{10} at all sites (Hitzenberger et al., 2006a; U.S.EPA, 2012b; Viidanoja et al., 2002; WHO, 2003). Its contribution to $PM_{2.5}$ increases to 15-20% at some curbside sites (Viidanoja et al., 2002; WHO, 2003). BC particles near sources, e.g. in fresh engine exhaust, are smaller than BC particles further away from sources (Bond et al., 2013; Smith et al., 2009). BC is carbonaceous and the most strongly light-absorbing component of particulate matter (U.S.EPA, 2012b).

BC is highly correlated with traffic-related pollutants like NO, NO_2 and ultrafine particles (Beckerman et al., 2008; Westerdahl et al., 2005). The correlation with $PM_{2.5}$ is much lower (Beckerman et al., 2008; Gan et al., 2011). These correlations do differ between different studies depending on the measurement location (close to traffic vs. background concentrations), and the geographical region.

Atmospheric lifetime of BC is around one week to a few weeks (Bond et al., 2013; Cape et al., 2012; Highwood and Kinnersley, 2006; Shindell et al., 2012). BC is removed from the atmosphere via precipitation and contact with surfaces / deposition (Bond et al., 2013).

2.4.2 MEASUREMENT OF BLACK CARBON

Despite efforts during the past decennia, no standard method exists to measure BC or EC. All current analysis methods are operationally defined: some use the optical properties or light-absorbing characteristics, others use the thermal and chemical stability of carbon (FIGURE 13) (U.S.EPA, 2012b).

Measurement of BC relies on the modification of the optical properties of a fiber filter matrix by deposited particles (Petzold et al., 2005; Quincey et al., 2009). Three methods can be distinguished: filter transmittance measurement, filter reflectance measurement, and a combination of transmittance and reflectance. The mass concentration is determined using a conversion factor from light absorbance to mass (Wallace et al., 2011). A BC reading can be calculated with a time resolution of up to 1 second. However, integration over longer time intervals is better as this increases the reliability of the measurement.

Frequently used devices that calculate the transmittance of a filter are the aethalometer and the particle soot absorption photometer (PSAP). These devices detect the changing optical absorption of light transmitted through a filter ticket. A multiangle absorption photometer (MAAP) takes into account transmitted and reflected light. Comparisons between aethalometers, PSAPs and MAAPs showed that methods agreed well, with R^2 values of over 0.9 (Hitzenberger et al., 2006b; Müller et al., 2011).

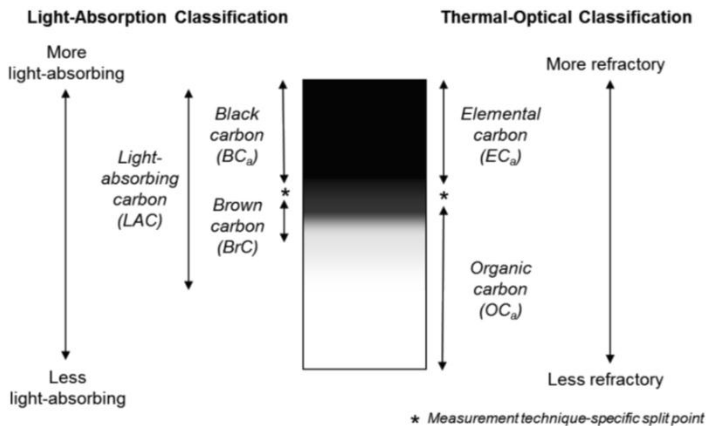


FIGURE 13: Measurement of the carbonaceous components of particles (U.S.EPA, 2012b)

EC is determined using a simple thermal or a thermal-optical method: particles are captured on a filter, and that filter is analyzed in a laboratory following different temperature protocols (Chi, 2009; Reisinger et al., 2008). The simple thermal method typically overestimates EC and underestimates OC because some organic compounds pyrolyze during the analysis (Chi, 2009). The thermal-optical technique is similar to the thermal technique, but the reflectance (TOR) and/or transmittance (TOT) of light is continuously monitored. A major disadvantage of the thermal and thermal-optical methods is the low temporal resolution of the measurements: one filter integrates concentrations over at least several hours. There are currently two common techniques employed for the analysis of elemental carbon and organic carbon (ECOC) in atmospheric particulate matter samples: the IMPROVE ECOC method, and the NIOSH ECOC method (Schauer, 2003). Currie et al. (2002) and Schmid et al. (2001) have presented a comprehensive comparison of these methods and a large number of other EC analysis methods. An alternative to the off-line laboratory-based analysis of samples, is the semi-continuous ECOC analyzer that uses a methodology comparable to the NIOSH method.

The analytical determination of EC is complex. It requires a non-carbon filter, the measurements are time-consuming, expensive, and filter destructing (Cyrys et al., 2003). Measuring the reflectance of PM filters is an alternative and cheaper way to assess exposure to traffic-related particulate matter. The absorbance of $PM_{2.5}$ ($PM_{2.5}$abs) can be determined by measuring the reflectance of filters used for mass determination, e.g. by using a Smoke Stain Reflectometer. Whereas BC and EC are determined as a mass ($\mu g * m^{-3}$), $PM_{2.5}$abs or reflectance is expressed in $10^{-5} * m^{-1}$.

Although BC, EC and $PM_{2.5}$abs are measured differently using optical and/or thermal methods, these pollutants are in most cases highly correlated (Cyrys et al., 2003; Schauer, 2003). From our own measurements in Flanders in two seasons, it appeared that BC $\cong$ 1.5 * EC, with an R^2 of >0.9 (EC determined by thermal-optical analysis using the NIOSH temperature protocol; and BC measured with a micro-aethalometer) (Van Poppel et al., 2012a). It is important to recognize that BC should not be used as a direct measure of EC, and that the levels of BC, EC, or $PM_{2.5}$abs measured in different studies using different measurement techniques are not directly comparable (Hitzenberger et al., 2006b; Schauer, 2003).

2.4.3 BLACK CARBON EMISSIONS AND DISPERSION

Emissions inventories list major sources of BC, both anthropogenic and natural, and the trends in BC emissions over time (U.S.EPA, 2012b). Understanding the sources of BC is needed for designing mitigation strategies capable of achieving both climate and public health benefits (UNECE, 2010).

In the past, BC emissions have increased almost linearly, totaling about 1000 Gg (approximately 1.1 million tons) in 1850, 2200 Gg (approximately 2.4 million tons) in 1900, 3000 Gg (approximately 3.3 million tons) in 1950, and 4400 Gg (approximately 4.8 million tons) in 2000 (Bond et al., 2007). However BC and OC inventories have an estimated uncertainty up to a factor of 2 (Bond et al., 2004); e.g. the U.S. EPA estimates the global BC emissions at 7600 Gg (8.4 million ton) for 2000 (U.S.EPA, 2012b). In Western Europe and North America, there has been a substantial decline of BC over the past decades, mainly because of a decrease in coal burning (Novakov and Hansen, 2004). In contrast, emissions of BC in developing countries (China, India, Latin America) have increased exponentially (Bond et al., 2007). The spatial distribution of these emissions shows Asia, parts of Africa, and parts of Latin America to be among the regions emitting the largest amounts of BC (75% of worldwide BC). Developed world regions like Europe, Japan, and the Middle East currently have lower BC emissions (U.S.EPA, 2012b).

In Western Europe, the most important source of BC is transport, responsible for 58% of the emissions (U.S.EPA, 2012b). Industry is responsible for 24% of BC emissions, residential and domestic sources for 11%. Open biomass burning, including wildfires and agricultural burning, is marginal in Europe (6%), but is a very important source in other regions of the world. Energy supply accounts for only 1% of emissions in Western Europe.

Incomplete combustion of fossil fuels is thus a major contributor in Western Europe (Kinney et al., 2000; Novakov and Hansen, 2004). Diesel cars emit lots of BC, although the implementation of emission controls, especially the introduction of (diesel) particle filters, are expected to reduce road vehicle exhaust emissions of BC by more than 80% and 95% until 2020 and 2030 respectively, and even further until 2050 (IIASA, 2012). Non-exhaust emissions (tire and brake wear, road abrasion) will be the dominant source of BC in the future as no controls are in place for these emissions (IIASA, 2012). Compared to other sectors, the importance of BC emissions from incomplete combustion of fossil fuels will decrease in the future.
Local sources of BC include smoking, candle burning, gas stove usage, and wood burning for residential heating (Lai et al., 2006; LaRosa et al., 2002).

Meteorology influences ambient BC concentrations, next to local and regional emissions of BC. Increasing wind speed and increasing humidity, leads to a decrease in ambient BC levels (Lai et al., 2006; LaRosa et al., 2002; Zagury et al., 2000). Temperature and mixing height also affect BC concentrations (Jung et al., 2010; Lai et al., 2006). Wind direction is important on the local scale.

Traffic is an important source of BC; moreover it is important when assessing personal exposure as these emissions occur close to people. BC or EC show a steep decline in concentrations when moving away from a road (Ducret-Stich et al., 2013; Karner et al., 2010; Padro-Martinez et al., 2012; Zhu et al., 2002). Karner et al. (2010) found that at a distance of 130m from a major road, BC concentrations have declined with 56%. These distance-decay gradients are much larger as compared to other pollutants like NO_2, $PM_{2.5}$ or PM_{10}.

Global ground-level BC measurements indicate estimated concentrations ranging from <0.1 µg/m³ in remote locations to ~15 µg/m³ in urban centers (U.S.EPA, 2012b). Ambient levels in China are approximately 10 times higher (in both urban and rural areas) than those in North America or Europe (U.S.EPA, 2012b).

2.4.4 HEALTH EFFECTS

In dedicated studies, it has been shown that traffic exposure may trigger health effects like myocardial infarction (Brook et al., 2010; Mills et al., 2007; Nawrot et al., 2011; Peters et al., 2004). BC or other traffic-related pollutants correlated with BC (NO_2, CO, EC, UFP), may also provoke short or longer term health effects. Health outcomes associated with BC include cardiovascular effects (Adar et al., 2007; McCracken et al., 2010; Wellenius et al., 2012), respiratory effects (Lin et al., 2011; Patel et al., 2010), neurological effects (Power et al., 2011; Suglia et al., 2008), and mortality (Gan et al., 2011). Some of these effects can be demonstrated at ambient concentrations below 1 µg/m^3 (Adar et al., 2007; Hoffmann et al., 2012; McCracken et al., 2010). There is not enough evidence to support a definitive causal association between exposure to BC (measured as EC) and increased risks for lung cancer (Gamble et al., 2012).

An increase in annual BC with 250 ng/m^3 was associated with a 7.6% decrease (95% CI, −12.8 to −2.1) in leukocyte telomere length (McCracken et al., 2010). Telomere length reflects biological age and is inversely associated with risk of cardiovascular disease. An IQR change in the 24-hour mean concentration (459 ng/m^3) results in a 19% decrease (95% CI, -21 to -17%) in high frequency power when studying heart rate variability (Adar et al., 2007). Baja et al. (2010) examined the effects of BC on heart-rate–corrected QT interval (QTc), an electrocardiographic marker of ventricular repolarization. An IQR change in BC cumulative during the 10 hours before the visit (550 ng/m^3) was associated with increased QTc (1.89 msec change; 95% CI: −0.16 to 3.93). Ischemic stroke is linked to an increase in BC pollution of 600 ng/m^3 24 hours preceding the stroke onset (OR (95% CI): 1.08 (1.01-1.16)) (Wellenius et al., 2012). Long term exposure to BC (IQR increase of 800 ng/m^3) was associated with a 3% increase in coronary heart disease (CHD) hospitalization and a 6% increase in CHD mortality (Gan et al., 2011). Zanobetti et al. (2006) found an 8.3% increase (95% CI, 0.2 to 15.8%) in the risk of emergency myocardial infarction hospitalization for an increase of 1700 ng/m^3 the day and the day before the admission, and a 11.7% (95% CI, 4.8 to 17.4%) increase in the risk of pneumonia hospitalization for a 1700 ng/m^3 increase in BC. Hoffmann et al.

(2012) found an absolute increase in systolic blood pressure of 2.2 mmHg (95% CI, 0.4 to 4.0 mmHG) with an increase in 5-day average BC (IQR = 250 ng/m^3). Mordukhovich et al. (2009) found similar results: a 7-day average increase in BC of 430 ng/m^3 was associated with an increase in systolic blood pressure of 1.5 mmHG (95% CI, 0.1 to 2.8 mmHg).

Patel et al. (2010) found that an increase in exposure to BC with 1200 ng/m^3 led to significant acute respiratory effects in adolescents. BC averaged over 24 hours was strongly associated with exhaled nitric oxide, an acute respiratory inflammation biomarker: a 16.6% increase (95% CI, 14.1 to 19.2%) per IQR increase in BC (4000 ng/m^3) (2011). Cyclists are exposed to higher levels of BC during commute compared to non-cyclists; airway macrophage carbon was significantly associated with monitored commute BC in 28 healthy adults (Nwokoro et al., 2012).

An IQR increase (400 ng/m^3) of exposure to BC decreased cognitive function across assessments of verbal and nonverbal intelligence and memory constructs (Suglia et al., 2008). The odds of having a low score in the Mini-Mental State Examination test was 1.3 times higher for each doubling in 1 year BC concentration in a cohort of older men (95% CI, 1.1 to 1.6) (Power et al., 2011). In the latter study BC exposure ranged from 30 ng/m^3 to 1770 ng/m^3.

Janssen et al. (2011) compared the health effects of $PM_{2.5}$ and BC (expressed as 'black carbon particles'), and the increase in life expectancy because of a decrease in $PM_{2.5}$ and BC concentrations. It was calculated that a reduction in $PM_{2.5}$ of 1 µg/m^3 was equivalent to a reduction in BC of 0.4-0.7 µg/m^3 (EC in roadside PM is estimated to be 40 to 70%). This reduction leads to a gain in life expectancy of 21 days when the effect of $PM_{2.5}$ is considered. In contrast, a reduction in BC of 0.4-0.7 µg/m^3 leads to a gain in life expectancy of 3.1 to 4.5 months. Janssen et al. (2011) concluded that the estimated increase in life expectancy associated with a hypothetical traffic abatement measure was four to nine times higher when expressed in BC compared to an equivalent change in $PM_{2.5}$ mass. BC is thus a useful additional indicator to evaluate health risks of airborne particles.

2.4.5 ENVIRONMENTAL EFFECTS

BC is not a greenhouse gas, but as a solid particle or aerosol it contributes to warming of the atmosphere. CO_2 is the most important human emission in terms of climate-forcing, but BC is estimated to come at a second place, before CH_4 and O_3 (Bond et al., 2013; Hansen et al., 2005). BC has a unique and important role in the Earth's climate system because it absorbs solar radiation, influences cloud processes, and alters the melting of snow and ice cover (Bond et al., 2013). The direct and snow/ice albedo effects of BC are widely understood to lead to climate warming (U.S.EPA, 2012b). The indirect effects of BC on climate via interaction with clouds are much more uncertain, but may partially offset the warming effects (U.S.EPA, 2012b). Concentrations respond quickly to reductions in emissions because BC is rapidly removed from the atmosphere by deposition: the atmospheric lifetime of BC is days to weeks, whereas CO_2 stays in the atmosphere for up to millennia (U.S.EPA, 2012b).
Under the 1997 Kyoto Protocol, no control of black carbon (BC) was considered (Jacobson, 2002). But BC emission reductions represent a potential mitigation strategy that could reduce global climate forcing from anthropogenic activities in the short term (due to the short lifetime of BC in the atmosphere) and slow the associated rate of climate change (Bond et al., 2013; Jacobson, 2002; Shindell et al., 2012).

2.4.6 EFFECTS ON BUILDINGS AND MATERIALS

An environmental impact close to sources in the urban environment is BC deposited on the lower levels of buildings (Highwood and Kinnersley, 2006; U.S.EPA, 2012b). Sýkorová et al. (2011) used benzonitrile as an indicator for BC in the analysis of materials, and it was shown that blackness of buildings is indeed caused by local BC emissions. In conclusion, deposition of BC darkens surfaces and has important aesthetic implications for buildings (U.S.EPA, 2012b).

2.4.7 CONCLUSIONS

- BC, EC or $PM_{2.5}$abs can be measured using thermal or optical methods. As there is no universally accepted standard, no method can be labeled as the 'correct' BC.
- Emissions from mobile sources are the dominant BC source in Western Europe.
- BC has steep distance-decay rates when moving away from roads: approximately 100m from a highway, concentrations are almost at background levels.
- BC emission control measures have important climate benefits, but they also have substantial co-benefits for air quality and public health worldwide (Anenberg et al., 2012).
- Health outcomes associated with BC include cardiovascular, respiratory, and neurological effects, and mortality. BC is also implicated in global warming.
- BC has gained more attention in recent years, resulting in its inclusion in several high level policy documents (U.S.EPA, 2012b; UNECE, 2010; UNEP, 2011; WHO, 2012).

3. MEASURING PERSONAL EXPOSURE

3.1 IMPACT OF TIME-ACTIVITY PATTERNS ON PERSONAL EXPOSURE TO BLACK CARBON

This chapter is based on:

Dons, E., Int Panis, L., Van Poppel, M., Theunis, J., Willems, H., Torfs, R., Wets, G., 2011. Impact of time-activity patterns on personal exposure to black carbon. Atmospheric Environment 45, 3594-3602.

3.1.1 INTRODUCTION

Personal exposure can be defined as the real exposure as it is experienced by individuals. When an individual is present at a certain place or in a certain microenvironment, he or she is exposed to the pollutant concentrations at this specific place. When an individual makes a trip from location A to location B, his personal exposure can be defined as the weighted average of concentrations present at each single location (WHO, 1999). Up till now, personal exposure is often estimated through the use of concentrations measured at fixed monitoring stations (Kaur et al., 2007; Sarnat et al., 2010). This is an approximation, as not only the ambient concentration is relevant, but also concentrations in different microenvironments (including indoors) and the whereabouts of individuals (Boudet et al., 2001; Jensen, 1999; Klepeis, 2006). Several studies have already examined the correlation between personal exposure and concentrations measured at fixed monitoring stations (Avery et al., 2010; Boudet et al., 2001; Gulliver and Briggs, 2004). This correlation shows a large spread between different studies, but overall correlation is stronger for longitudinal within-person studies, compared to cross-sectional studies (Avery et al., 2010). This indicates that differences between people and a large part of the spread within a subject can be explained by the activity pattern of the individuals and their daily environment.

Several studies are looking at the relationship between levels of exposure and health effects, but epidemiologists experience vast problems with exactly quantifying exposure. 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; Setton et al., 2011). Therefore researchers are looking at methods, either through direct measurements or indirect modeling, to reduce exposure misclassification (Int Panis, 2010).

We hypothesize that people, who are living at the same location, can nevertheless have a different exposure profile. The driving force for this difference will be the activity pattern and the subsequent microenvironments visited during a day. Short term exposures may contribute significantly to daily average exposure. The aim of this study is to look at week-long exposure profiles with a high temporal resolution. Linking these data with detailed time-activity patterns will tell us what the impact is of an activity pattern on personal exposure. Two groups of people with a highly differential time-activity pattern were selected to demonstrate this.

The pollutant looked at is black carbon (BC). BC has been used as an indicator of exposure to diesel exhaust (HEI, 2010), and it has been suspected as a contributor to global warming (Highwood and Kinnersley, 2006). Several researchers have recently stressed potential short and long term cardiovascular, respiratory and neurodegenerative health effects of BC (Baja et al., 2010; McCracken et al., 2010; Patel et al., 2010; Suglia et al., 2008). Over the last 40 years BC concentrations have declined rapidly in Europe, although the air has still moderate to heavy BC pollution. In the last decade concentrations seem to have leveled off possibly because of increasing emissions of diesel passenger cars.

3.1.2 MATERIALS AND METHODS

3.1.2.1 Study design and sampling method

Personal exposure measurements were performed in Belgium from May 2nd to July 8th 2010. 16 participants were asked to carry a device to measure BC-concentrations and to record their activities and whereabouts in an electronic diary. The study population comprises 8 couples, consisting of a full-time worker and a homemaker. Participants performed their regular activities; there were no restrictions but weeks where respondents were abroad or planned a weekend trip were excluded. All participants had to be nonsmokers. The presence of children and several characteristics of the residence were recorded, but they were not exclusion criteria. Each couple was measured sequentially for a 7-day period. Since the devices had to be reconfigured after each use, at least one day was in between the measurements of two couples, preventing reoccurring potential bias towards the end of the week (e.g. less accurate registration of activities (Bellemans et al., 2007)). In addition to the two personal measurements, a third measurement device was installed outside in front of the house of the couple, at the street side, to measure outdoor concentrations simultaneously. Two couples lived in an urban environment, two in a suburban zone and four couples were living in a more rural area.

An aethalometer - microAeth Model AE51 (AethLabs, 2011) was used for personal monitoring of BC. This monitor is small and portable (250g) and has a battery autonomy of up to 24 hours when logging on a 5 minute basis, as in this study. Inside is a small teflon-coated borosilicate glass fiber filter where BC-particles are captured. The aethalometer detects the changing optical absorption of light transmitted through this filter ticket. Every two days participants were asked to replace the filter to prevent saturation and to maintain measurement integrity. The pump speed was initialized at a rate of 100 ml per minute. One of the aethalometers was used for outdoor measurements at the place of residence. For this purpose a weather-proof housing was developed. For the personal measurements, participants could carry the aethalometer in their own backpack or handbag. Specific attention was drawn to the fact that the tube connected to the pump always had to be exposed to the air.

Activities, trips and GPS-logs were recorded on a small handheld computer or PDA. PARROTS (PDA system for Activity Registration and Recording of Travel Scheduling) was developed to facilitate this process and to minimize respondent burden (Bellemans et al., 2008). This tool was already deployed in a large scale survey on 2,500 households, and a comparison was made with the traditional paper-and-pencil method. The electronic diary enforces all attributes of executed activities to be provided. Accordingly it resulted in a non-response of 0 and provides several consistency checks. It was concluded that PARROTS provides high quality time-activity data while no additional respondent attrition was observed (Kochan et al., 2010). A disadvantage is the limited battery autonomy of this device (approximately 4 hours), implying the need to recharge the PDA at the workplace, at a friend's residence or in the car, since a car charger was provided as well.

Participants to this study were instructed to report each activity by picking one of thirteen predefined categories. In addition, they had to indicate the date, the start and end times, and the location (choosing from 4 predefined categories). Start and end times were expected to be precise on a 5 minute time base. For trips, each respondent had to specify the start and end location and time, and the transport mode(s) used (choosing from 12 predefined categories). Finally, respondents had to indicate with whom they were performing an activity or trip.

In addition to the PDA and the aethalometers, a short questionnaire was handed over to each couple at the start of the measurements. Personal and household characteristics, e.g. birth year or car ownership, and housing characteristics, like the presence of air conditioning or location of the home next to a busy street, were asked to get an idea of possible confounders.

All participants were personally instructed on the aim of the study, how to use the devices and redirected to a help desk in case of problems during the week. No financial compensation was rewarded, but afterwards everyone received a personalized report.

3.1.2.2 Quality assurance

Three aethalometers were used during the study. For the comparison of the different devices, they were put next to each other for over a week to test their correspondence. We measured in the relevant range (0 - 10,000 ng/m^3) in a real life situation, including indoor measurements and near transport. Correlation of the three devices was very high ($r > 0.96$). Further BC concentrations were compared with a fixed monitoring station using the MAAP technique (Multi-Angle Absorption Photometry; station 42R801 Borgerhout, urban location), which was used as a reference value. Concentrations measured at the monitoring station AL01 (Antwerpen-Linkeroever) were considered as background concentrations.

When both partners were at home, they were asked to put the two personal aethalometers next to each other in the living room. In that way at least 7 hours of common measurements were available for each day. Consequently we could do a daily check on the accuracy of these two aethalometers. This resulted in a Pearson correlation coefficient of 0.92, not knowing for sure that participants accurately followed our instructions.
The accuracy of the recorded activities and trips was checked by consulting the GPS-logs. The diary of the partner was used to check for uniformity (e.g. if one person indicated that he was doing an activity with his/her partner, it had to be present in the other diary as well). If an inconsistency was detected, the participants were contacted shortly after the measurement period and asked to clarify the situation.

3.1.2.3 Data analysis

All devices, the three aethalometers and the two PDA's, were synchronized at the start of each week. Activities, trips, GPS-logs and BC-concentrations were directly loaded into a database to minimize manual work and counter possible introduction of errors.

Negative measurements were included into the analyses (McBean and Rovers, 1998). Because the aethalometer detects the change in optical absorption, small shifts in the light beam or the filter ticket can cause a temporary decrease in measured absorption. Since the aethalometer computes the difference with the previous measurement, negative measurements are offset in the next observation(s). Missing values were caused by low battery events or when devices were intentionally switched off for changing the filter ticket. We did not try to predict a value but rather kept the missing values and treated them as blanks. When directly comparing a full-time worker and a homemaker, we only used data for which simultaneous measurements were available. Aethalometers are suitable for personal measurements, but when switching from one microenvironment to another with different environmental conditions an adjustment effect can be observed (Wallace et al., 2011). This effect was observed as well but to a lesser extent than in previous studies since a larger integration time was used. A sensitivity check excluding all first observations before and after switching to a different microenvironment changed the average concentrations by 5%, showing the limited impact of this effect.

For the personal measurements, a total of 32,320 observations on a 5-minute timescale were conducted; 1352 values were missing. Of all valid observations, 460 measurements or 1.5% were negative. An overall mean concentration of 1578 ng/m^3, a median of 1108 ng/m^3 and a standard deviation of 2571 ng/m^3 are observed. There were 14,656 observations from the fixed outdoor aethalometers at the homes of the couples, 10.9% were missing and 2.1% were negative. The mean concentration is 1323 ng/m^3 and the median is 1112 ng/m^3.

Statistical analysis was performed in SAS 9.2.

3.1.3 RESULTS

3.1.3.1 Questionnaire data and time-activity patterns

The age of all sixteen participants was between 20 and 60 (since we recruited in the working population). Eight were male and eight female, with a small bias towards higher education. Everyone was in the possession of a driver's license. One household had no private car; other households had either one or two cars, all diesel.

TABLE 3 shows the percentage of time spent by the participants on each activity. The initial 13 activities, plus ‘in transport’, are grouped in eight broader categories. The largest amount of time is spent at the home location (sleeping, home-based activities), followed by working, social and leisure activities, and time spent in transport. These results are similar to results from a time-use survey held in Belgium in 2005 among 6400 respondents (FPS Economy - Statistics Division, 2008). The latter shows 71% of the activities at the home location, 10% of time is spent at work and 6% of time in transport.

Full-time workers spend more time in transport (112 minutes) than people without a full-time job (67 minutes). Homemakers were, as expected, more at home. The activity ‘Work’ is also observed for homemakers; this is explained by the fact that half of these men and women worked part-time.

TABLE 3: Time spent on each activity and exposure to BC during each activity in the personal exposure measurement campaign. Only periods when both partners had measurements, are included in this table to enable direct comparison between partners.

	Total				Full-time worker				Homemaker			
	Total # of 5 minute observa-tions (N)	Average time (%)	Average exposure (%)	Average exposure (ng/m^3)	# of 5 minute observa-tions (N)	Average time full-time worker (%)	Average exposure full-time worker (%)	Average exposure full-time worker (ng/m^3)	# of 5 minute observa-tions (N)	Average time home-maker (%)	Average exposure home-maker (%)	Average exposure home-maker (ng/m^3)
Sleep	10,654	34.3	25.0	1153	5154	33.1	23.3	1195	5500	35.4	26.9	1114
Home-based activities	9855	31.7	25.0	1223	4142	26.6	19.1	1192	5713	36.7	31.6	1246
Work	4278	13.8	10.8	1276	3318	21.3	14.8	1219	960	6.2	6.3	1454
Social and leisure	2728	8.8	7.4	1525	1087	7.0	6.1	1728	1641	10.6	9.0	1400
In trans-port	1933	6.2	25.6	6445	1206	7.8	31.5	6812	727	4.7	19.0	5858
Other	964	3.1	3.0	1495	380	2.4	2.4	1661	584	3.8	3.6	1393
Shopping	458	1.5	2.5	2584	141	0.9	2.0	3486	317	2.0	3.1	2183
Go for a ride	234	0.8	0.7	1723	124	0.8	0.9	2107	110	0.7	0.6	1282

3.1.3.2 Personal exposure measurements

TABLE 4 presents the average personal exposure of all 16 participants over a 7-day period. There are differences between the households and between members of the same household. The maximum difference between personal exposure of a full-time worker versus a homemaker of the same household amounts to 30%. In most cases the full-time worker is more exposed. The 7-day averages are more variable between weeks/locations than between members of the same household.

TABLE 4: Average personal BC-exposure, outdoor concentration measured at the home location of 8 households and average background concentration (ng/m³). The outdoor measurements for household 1 are limited due to a technical failure of the aethalometer.

	Home location	Average outdoor concentration at home (ng/m^3)	Average background concentration at fixed monitor [c] (ng/m^3)	Average exposure full-time worker (ng/m^3)	Average exposure homemaker (ng/m^3)	Difference between full-time worker and homemaker
HH1	Suburban	1160 [a]	960	1465	1023	30%
HH2	Urban	2138	1003	2079	1869	10%
HH3	Urban	1694	1459	2071	1750	15%
HH4	Rural	1313	1183	1428	1530	-7%
HH5	Rural	1367	1559	2130	1830	14%
HH6	Suburban	611 [b]	679	885	773	13%
HH7	Rural	1130	2020	1929	1413	27%
HH8	Rural	1200	1400	1580	1582	0%

[a] N=561
[b] at the back of the house
[c] fixed monitoring site AL01 (Antwerpen-Linkeroever)

A typical daily pattern of a couple, living in an urban area, is shown in FIGURE 14. A first peak for the full-time worker is while commuting by car; in the evening another peak is observed during his return home, this time using a slightly different route (according to the GPS). During the day concentrations at the workplace, located in a rural area, are lower than concentrations at his home location. His wife is at home till the early afternoon, when she leaves by bike for a social activity (trip takes approximately 15 minutes). After returning home, she stays at home for about an hour and leaves again for a leisure activity, again by bike. From 8.30 p.m. onwards, both man and woman are at home.

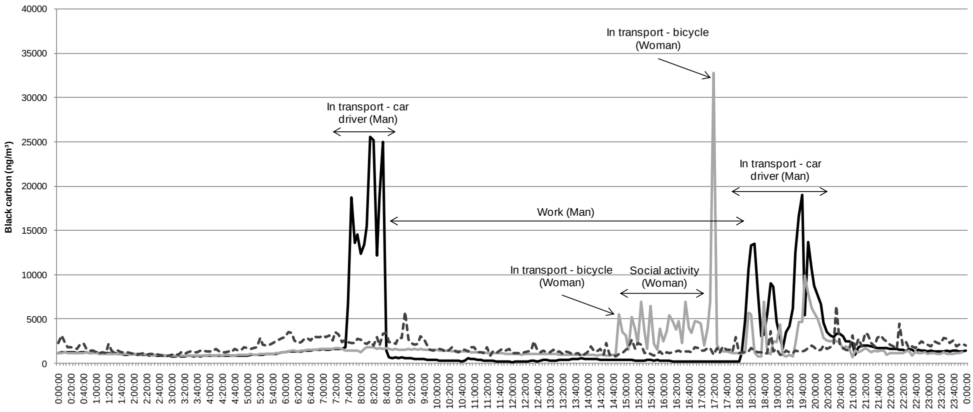


FIGURE 14: Personal (homemaker (gray) and full-time worker (black)) and outdoor (at the front of the house (dashed line)) concentrations on May 19, 2010

Differences in exposure between members of a family originate from differences between their time-activity pattern and the corresponding locations visited. When comparing exposure during different activities, BC concentrations vary substantially, both within one activity-category, and between different activities (FIGURE 15). Mean and median concentrations are higher in transport in comparison with all other activities; standard deviation is also largest in transport. Short activities (shopping, bring/get goods/people) may give slightly higher concentrations than in reality because of the difficulty in distinguishing those brief activities from the preceding or following transport activity. Participants had to report executed activities on a 5 minute basis but some short activities will not fit into this time period. Social and leisure activities capture a wide variety in meanings and locations, in accordance there is a broad range in concentrations with important peaks during café visits. Lowest concentrations are observed during home-based activities and at night, when respondents are sleeping.

FIGURE 15 also shows concentrations per location. Again it is very clear that concentrations in transport are highest. Concentrations in private homes are lowest, both in the residence of the participants as well as in residences from friends or family; although these differences are not significant.

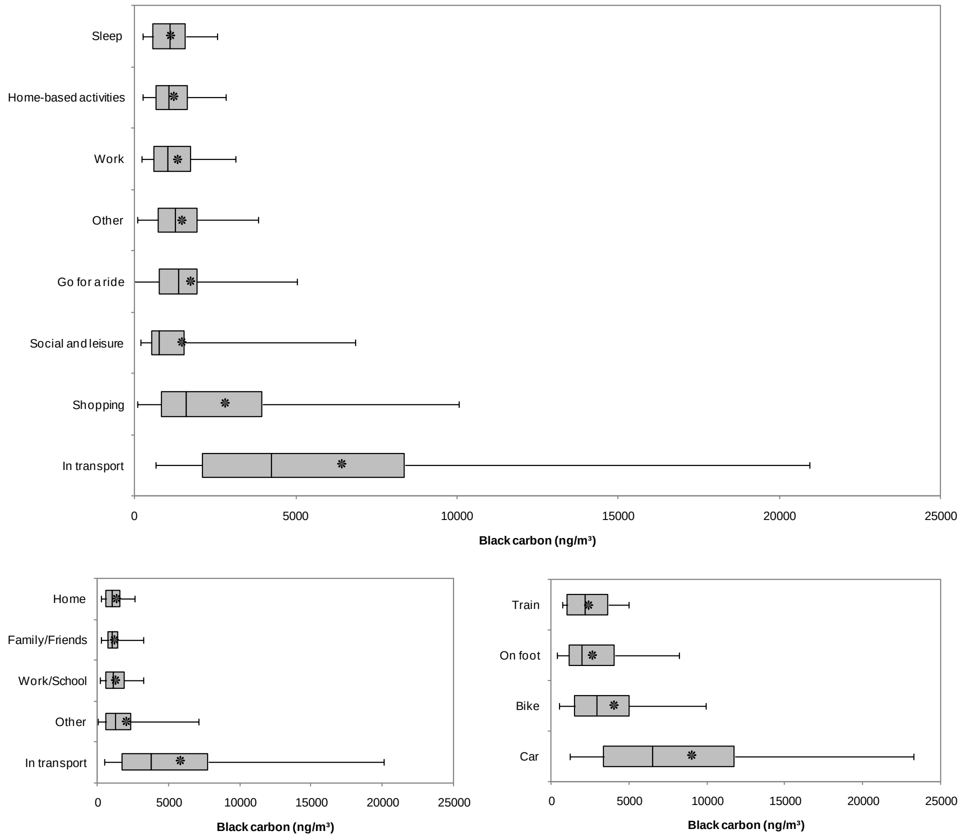


FIGURE 15: Concentrations measured per activity (upper), per location (under, left) and per transport mode (under, right). Represented are P5, 1st quartile, median, 3rd quartile and P95. The asterisk mark shows the mean value. Categories with less than 100 observations are omitted.

Concentrations in transport are higher than during any other activity, mainly caused by high exposure during car trips (both car driver and car passenger). When traveling by car, in-vehicle concentrations are on average 5 to 8 times higher than the average exposure at home and 4 to 7 times the average outdoor concentration at home. Concentrations on a train, and during walking or cycling trips are substantially lower, but still a factor 2 higher than the average concentration at the home location. A division can be made between concentrations during a functional and a recreational trip. Concentrations during a recreational bike trip (mean=1381 ng/m^3, N=174) are substantially lower than during a functional bike trip (mean=3674 ng/m^3, N=476).

When combining the exposure per activity with the time-activity data from the diaries, transport is the most important contributor to personal exposure (TABLE 3, FIGURE 16). Although people spend only a modest amount of time in transport (6.2% or 89 minutes per day), this is responsible for a quarter of total exposure to BC. When sleeping, the exposure/time ratio is lowest.

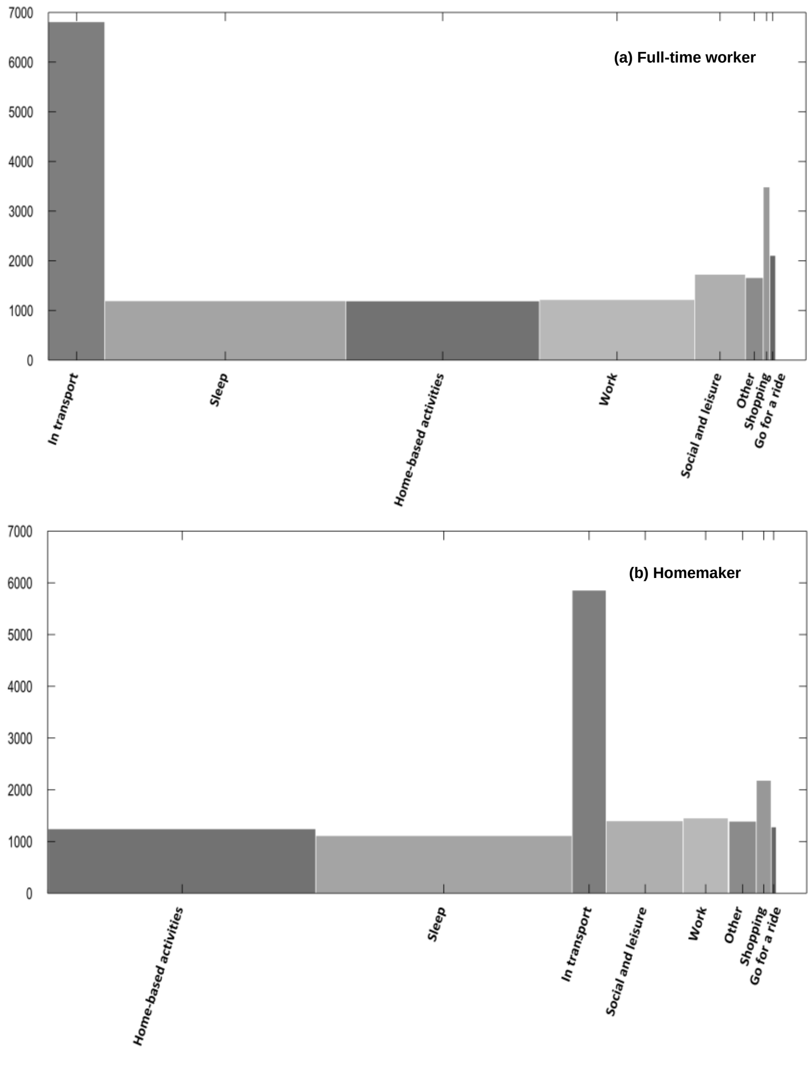


FIGURE 16: Average black carbon concentration (ng/m³) per activity is represented on the y-axis; average time spent doing a particular activity is represented on the x-axis. The area of the blocks signifies the total contribution of each activity to the personal accumulated exposure to BC. Blocks are arranged from left to right according to their surface area.

The largest contribution to total exposure for full-time workers is by far 'being in transport', due to the larger fraction of time in transport. For homemakers the contribution of exposure at home is more important.

Since some homemakers worked part-time, the difference in activity pattern with full-time workers may be blurred. For that reason a differentiation was made between working days, weekdays and weekends (TABLE 5). Working days were defined as 24h-periods where approximately 8 hours are spent on paid work, weekdays are Mondays till Fridays with no paid work at all, and weekends are all Saturdays, Sundays and public holidays. Average exposure on a working day is 24% higher than on a weekday. Comparing the 50th percentile and the 95th percentile of a working day and a weekday clearly shows that the 5 to 10 % highest values present on a working day (mainly caused by commuting trips) explain this difference. From the fixed monitoring network a difference between weekends and weekdays of about 20% was observed, with lower concentrations during the weekend.

TABLE 5: Average outdoor concentration (measured with the aethalometer at the front of each family's house), personal exposure, percentiles of personal exposure and average time in transport

	Average outdoor concentration (ng/m^3)	Average exposure (ng/m^3)	P5 (ng/m^3)	P10 (ng/m^3)	P25 (ng/m^3)	P50 (ng/m^3)	P75 (ng/m^3)	P90 (ng/m^3)	P95 (ng/m^3)	Average time in transport (#min in 24h)
Working day[a]	1338	1793	240	333	609	<u>1132</u>	1789	3021	<u>5140</u>	131 min
Weekday [b]		1366	259	367	637	<u>1042</u>	1629	2410	<u>3331</u>	60 min
Weekend[c]	1289	1527	182	306	642	1153	1782	2749	3917	69 min

[a] Working day = Mon/Tue/Wed/Thu/Fri (person works for 8h – as a profession)
[b] Weekday = Mon/Tue/Wed/Thu/Fri (person does not work)
[c] Weekend = Sat/Sun or public holiday

Differences between households are larger than differences between partners, as shown in TABLE 4. Both background concentrations and urban/rural location will thus have a greater impact on personal exposure than the activity pattern. Households 2 and 3 live in a densely populated urban residential area with rather low traffic intensities on the nearest street (less than 5000 vehicles per day); this suggests a higher personal exposure when living in a city compared to more suburban or rural locations. The low values for household 6, living in a suburban region, are to a large extent explained by the low background values measured during that period at fixed monitoring stations.

3.1.3.3 Outdoor measurements

The dotted line in FIGURE 14 represents the concentrations measured by the fixed outdoor aethalometer at the front of the house. Outdoor concentrations show little variation, although in the morning concentrations are somewhat higher. This trend can be observed at all locations and in all eight measurement periods, most probably due to the relatively low traffic intensities at the home addresses of the participants in this study. In TABLE 5 a distinction is made between weekdays and weekends, showing higher concentrations on weekdays. Urban outdoor concentrations are higher than concentrations in suburban or rural areas, while during this period background concentrations are not significantly higher than during other weeks (TABLE 4). Average outdoor concentrations are lower than personal exposures, except for household 2. This can be explained by the location of this home in a dense urban area (urban background), while the inhabitants move out of the urban area to work and do leisure in less polluted areas.

An indoor-outdoor infiltration factor was calculated at times when partners were both at home; so we could compare indoor measurements with the outdoor monitor at the front of the house. The Pearson correlation between indoor and outdoor measurements was 0.66; (unidentified) indoor sources lower this correlation (FIGURE 17). This coefficient differs from residence to residence, ranging from 0.1 to 0.87. The slope of the regression function is positive but for every household smaller than 1, meaning that indoor concentrations are on average lower than outdoor concentrations. Correlation is higher for daily average indoor versus outdoor concentrations (r = 0.89). Overall we can conclude that higher outdoor concentrations correspond to increased indoor concentrations.

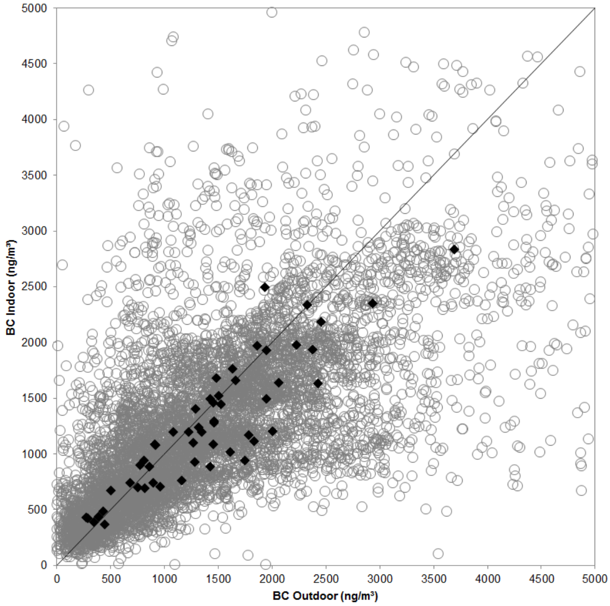


FIGURE 17: Calculation of infiltration factor at the 8 homes of participating families, based on indoor and outdoor observations between 0 and 5,000 ng/m^3. Indoor concentration is calculated based on the average of both personal aethalometers. Pearson correlation of all 5-minute observations is 0.66 (in gray); Pearson correlation of daily average indoor and outdoor concentrations is 0.89 (in black).

3.1.4 DISCUSSION

For BC, our personal monitoring study did reveal an undeniable contribution from the transport microenvironment. The amount of time in transport and the transport mode are important determinants of personal exposure to BC. People living at the same location and in the same residence, as the couples in this study did, sometimes had a completely different exposure, largely explained by the difference in activity pattern and their corresponding time in transport. This confirms earlier studies on the relationship between activities and air pollution that suggested a possibly important role for the traffic microenvironment, in contrast with the limited time spent in an outdoor or a traffic environment (Beckx et al., 2009b; Fruin et al., 2004). Time spent in or near transport may thus provoke a dissimilarity in personal exposure between 2 individuals living at the same location. When this can be demonstrated for a traffic-related pollutant like BC, it is most probably also the case for other pollutants that are highly correlated with BC or highly correlated with traffic. NO_2, soot and ultrafine particles show a good correlation with BC; correlation with PM_{10} and $PM_{2.5}$ is rather low (Beckerman et al., 2008; Berghmans et al., 2009; Hagler et al., 2009; Hoek et al., 2008; Westerdahl et al., 2005).

In-vehicle concentrations are higher than concentrations experienced on the bicycle, on foot or in a train. In a study of Fruin et al. (2004) the average in-vehicle BC exposure was 4100 ng/m^3. The large amount of diesel cars in Belgium (over 60% of all private cars are diesel (NIS, 2010)) is most likely responsible for the higher in-vehicle concentrations found in this study.
Variations in concentrations in one transport mode and between transport modes will not be explained in detail, as this was not the aim of this study, but can be, at least partly, addressed when analyzing the GPS-tracks. Short term peaks in transport are more prevalent during bicycle trips than during car trips where levels are more smoothly elevated; this was also observed by Int Panis et al. (2010) and Zuurbier et al. (2010). Shorter peaks, e.g. caused by a passing truck, will be spread over a 5 minute period. It is unclear to what extent short term exposure is relevant for health: it might be the high peaks that cause health effects or the longer periods of exposure to elevated levels or a combination of both (de Hartog et al., 2010a; Int Panis et al., 2010; Jacobs et al., 2010; Peters et al., 2004). When analyzing the difference between a functional and a recreational bike trip, exposure in the latter is remarkably lower (the diary makes a distinction between a functional trip and 'going for a ride'). This difference is probably due to the choice of route: As can be seen from Hertel et al. (2008) and Int Panis et al. (2010) proper choice of route can significantly lower exposure. The same conclusion holds for trips on foot although the difference is less pronounced.

Exposure in microenvironments different from the transport microenvironment is measured in several dedicated studies for a number of pollutants (Brown et al., 2009a; Cyrys et al., 2010; McConnell et al., 2010; Wallace and Ott, 2011). A lot of different classifications of microenvironments have been used in the past. In this study 5 broader categories were picked: home, work/school, in transport, house of family/friends, other. A disadvantage of this classification is the inability to make a distinction between indoor and outdoor microenvironments. However, we know from the questionnaires that all full-time workers were employed in an indoor environment. In most studies on air quality in specific microenvironments only a few different shops, restaurants or homes could be measured because of the limited number of measurement devices. In this study concentrations in several different locations were measured, namely when participants were visiting these places. Higher than average concentrations were observed in shops, both for daily (food, newspaper,…) as for non-daily shopping (clothes, furniture,…). Concentrations in shops are still double the concentration in homes after the removal of the first and last 5 minute-observation. This sensitivity check was necessary because of our suspicion that transport was partly included in these typically short activities and because of the adjustment effect of aethalometers to new environmental conditions (Wallace et al., 2011). The lowest concentrations were observed inside homes. Indoor peaks at home mostly originate from candle burning or woodstoves, but since this is rarely done in summer, indoor concentrations are primarily influenced by ambient pollution that infiltrates in the residence (Lai et al., 2006; LaRosa et al., 2002).

Suspicion of health effects is the main reason for calculating or measuring exposure. The more exact personal exposure can be determined, the less exposure misclassification will occur. Since we demonstrated that the maximum difference between 2 individuals living at the same location can differ by as much as 30%, using modeled or measured concentrations at the place of residence alone is neither accurate nor sufficient. Secondly, with the aim of calculating dose-response functions, it is necessary not only to calculate exposure, but also to correctly determine inhaled air, and subsequently to derive health effects for a specific dose. The fraction of inhaled air varies from person to person (e.g. influence of sex and age). But also performing certain (physical)

activities can affect breathing rates (Marshall et al., 2006; McConnell et al., 2010). The importance of taking into account breathing rates, especially while in transport, is demonstrated by Int Panis et al. (2010) and Zuurbier et al. (2010). With the study design as it is here, it's relatively straightforward to relate breathing rates as stated in international literature, to one of 13 activities or to a transport mode.
Epidemiologic studies relate BC exposure to cardiovascular, respiratory and neurodegenerative effects. An increase in annual BC with 250 ng/m^3 was associated with a 7.6% decrease (95% confidence interval, −12.8 to −2.1) in leukocyte telomere length (McCracken et al., 2010). Telomere length reflects biological age and is inversely associated with risk of cardiovascular disease. Baja et al. (2010) examined the effects of BC on heart-rate–corrected QT interval (QTc), an electrocardiographic marker of ventricular repolarization. An interquartile-range change in BC cumulative during the 10 hr before the visit (550 ng/m^3) was associated with increased QTc (1.89 msec change; 95% confidence interval: −0.16 to 3.93). Patel et al. (2010) found that an increase in exposure to BC with 1200 ng/m^3 led to significant acute respiratory effects in adolescents. An interquartile-range increase (400 ng/m^3) of exposure to BC decreased cognitive function across assessments of verbal and nonverbal intelligence and memory constructs (Suglia et al., 2008). An average difference between partners of 251 ng/m^3, as observed in this study, can thus be relevant for health.

It should be noted that we only did measurements in summer. Results need to be confirmed in other seasons. In a Californian study it is shown that monthly averaged BC-concentrations can be up to five times greater in winter than summer (Kirchstetter et al., 2008). Also in a European context concentrations in winter are greater than in summer (Adams et al., 2002; Kaur et al., 2007). Concentrations measured in weekends tend to be lower than concentrations on weekdays, consistent with the lower number of diesel trucks on the road (Kirchstetter et al., 2008). A weekly cycle is apparent in Belgium as well, both from our own outdoor measurements as from the fixed monitoring network. As concentrations in ambient levels vary over time, this will most likely have an effect on personal exposure as well. The design can be improved further by deploying more instruments and measuring multiple families in urban and rural locations at the same time.

To conclude, we can state that despite the important differences in background concentrations from week to week and the sequential measurement strategy, activities, microenvironments and transport modes with higher than average exposure can still be identified. Outdoor and personal exposure of men and women from the same household can be compared directly, since we measured simultaneously with the same equipment, thus ruling out potential bias related to temporal variation and sampling method. We can conclude that for certain households a considerable difference of up to 30% exists between both partners. This can be partially explained by the time in transport, and thus by the time-activity pattern. Differences between households are to a large extent attributable to changing background concentrations (as a consequence of our sequential measurement strategy) and to the location of the residence in an urban, suburban or rural environment.

Many models are built that use observed activity patterns or time-use data, e.g. activity patterns from NHAPS (National Human Activity Pattern Survey). Those activity patterns mostly lack the exact location (address, coordinate, municipality, etc.) where a specific activity is executed. That's why it is very difficult to link these patterns to air pollution concentrations with a high spatial and/or temporal resolution. The importance of detailed modeling of trips is underlined by this research, as we demonstrated that transport contributes significantly to total accumulated exposure. Activity-based models seem very promising in this area (Beckx et al., 2009a; Beckx et al., 2009c; Hatzopoulou and Miller, 2010; Marshall et al., 2006; Recker and Parimi, 1999; Shiftan, 2000). An important advantage of these models is the emphasis on traffic, since these models originate from traffic science. Once personal exposure is modeled, validation of the modeling framework can be done by a personal monitoring study as described here.

3.2 PERSONAL EXPOSURE TO BLACK CARBON IN TRANSPORT MICROENVIRONMENTS

This chapter is based on:

Dons, E., Int Panis, L., Van Poppel, M., Theunis, J., Wets, G., 2012. Personal exposure to black carbon in transport microenvironments. Atmospheric Environment 55, 392-398.

3.2.1 INTRODUCTION

In dedicated studies, it has been shown that traffic exposure may trigger health effects like myocardial infarction (Brook et al., 2010; Mills et al., 2007; Nawrot et al., 2011; Peters et al., 2004). Black carbon (BC), or other traffic-related pollutants correlated with BC (NO_2, CO, Elemental Carbon, Ultrafine particles), may also provoke short or longer term health effects, e.g. cardiovascular disease (Baja et al., 2010; Gan et al., 2011; McCracken et al., 2010), adverse respiratory health outcomes (Lin et al., 2011; McCreanor et al., 2007; Patel et al., 2010) or neurological effects (Bos et al., 2011; Power et al., 2011; Suglia et al., 2008). Recently it has been stressed by Janssen et al. (2011) that BC is a useful new indicator for the adverse health effect of traffic-related air pollution.

Typically epidemiological studies try to relate an exposure metric to certain health effects in exposed or less exposed people. If using a generic exposure metric like population exposure or air quality measured at one specific place, it neglects the large contrast and variation in personal exposures that is important in epidemiological studies. For example, individuals travelling from hot spot to hot spot or professional drivers will be exposed to far higher concentrations compared to a hypothetical static population. In previous studies using activity-based models or personal monitors it is demonstrated that the transport activity, although short in duration, can be responsible for quite a large part of integrated personal exposure to combustion-related pollutants (Beckx et al., 2009b; Dons et al., 2011b; Fruin et al., 2004; Marshall et al., 2006). Understanding the variation in exposure can contribute to a more accurate exposure assessment and reduce misclassification of air pollution health effects. This is of major importance when trying to define the health effects of pollutants that are highly variable in time and space, like e.g. traffic-related air pollutants (Setton et al., 2011; Strickland et al., 2011; Van Roosbroeck et al., 2008).

In the light of understanding the role of transport activities on total accumulated exposure, a large personal monitoring campaign was set up. BC was measured on a 5-minute time resolution, allowing air quality data to be linked with reported activities. In this paper the focus will be on exposure in traffic microenvironments; however transport is always considered as part of a complete 24h diary, enabling the identification of trip motives and the calculation of the contribution of transport to integrated exposure and inhaled dose. BC was chosen as a pollutant because of its relevance for health, and because of the availability of suitable measurement devices. Moreover the interest of policy makers in BC was aroused due to emerging evidence on health effects and the impact of BC on global warming. In developed countries, motorized transport, and mainly diesel vehicles, are considered to be the most important source of BC, whereas in developing countries biomass burning may be important (Highwood and Kinnersley, 2006; Kirchstetter et al., 2008).

3.2.2 MATERIALS AND METHODS

Personal exposure to BC is measured with portable aethalometers (microAeth Model AE51, (AethLabs, 2011)), carried by 62 individuals for 7 consecutive days. During the sampling, participants were urged to meticulously keep track of their executed activities by reporting them in an electronic diary fitted with a GPS. On top of that, a short questionnaire asked for characteristics of the individual, the household and the residence. More details on the configuration of the devices, quality assurance and data analysis can be found in chapter 3.1. Sixteen people took part in a pilot study in summer 2010; half of them participated again in a more elaborate campaign in winter 2010-2011. The other half was either unwilling or unable to participate a second time. The winter campaign was supplemented with 38 new volunteers. Because we wanted to focus primarily on the impact of the time-activity pattern on personal exposure, we measured two people sharing the same residence. In summer 2010 all 8 couples were measured sequentially; in the winter campaign a maximum of three couples were measured simultaneously each week, for eight weeks in a row.

Some small adaptations were made in the winter campaign compared to the summer. The PARROTS software installed on a small handheld computer (Kochan et al., 2010), to fill in executed activities and trips, was somewhat simplified to further reduce respondent burden, without significant data loss. In the first campaign couples consisted of a full-time worker and a homemaker or part-time worker; in winter we relaxed this constraint: there was no further limitation on the work schedule. Other adaptations all concerned quality assurance and quality control (e.g. additional comparison with filter-based EC analysis and with a Multi-Angle Absorption Photometer (MAAP) measuring BC simultaneously at the official air quality monitoring stations).

To maintain data integrity, we corrected the aethalometer readings in different ways. First, all data showing an error code were excluded from the dataset (except for low battery events). In addition, we excluded data when the attenuation was above 75, whereas the instrument only gives an error code if attenuation is around 100. The value of 75 is a conservative lower limit as proposed by Virkkula et al. (2007); Hansen (2005) proposed the range of 75 to 125 as a suitable advisory limit for aethalometers. Finally we did an intercomparison between all 13 devices used, to correct for device specific deviations (appendix FIGURE A1, corrections were between 1% and 23%). The sample flow of all instruments, set at 100 ml/min, was checked before the measurement campaign.

During the sampling campaign, data from a fixed BC monitor on a suburban background location (station 40AL01 – Antwerpen Linkeroever, operated by the Flemish Environment Agency) was used to correct for non-simultaneous measurements (for the methodology, see appendix A.1).

Negative measurements were included into the analysis, because a temporary false decrease in measured absorption is offset in the next observation(s) (McBean and Rovers, 1998; Wallace, 2005). Only deleting the negative values would overestimate average BC concentrations.

To calculate the contribution of each activity to dose, a translation of exposure to inhaled dose is made by defining a minute ventilation per activity and per transport mode; gender was also taken into account. Inhalation rates are based on Allan and Richardson (1998) and Int Panis et al. (2010) (appendix TABLE A2).

SAS 9.2 was used for data processing and statistical analysis.

3.2.3 RESULTS

3.2.3.1 Study characteristics and time-activity patterns

All 62 volunteers participating in the measurement campaign measured their personal exposure for 7 consecutive days, 24 hours a day, on a 5-minute time resolution. This resulted in 124,992 single measurements, or more than 10,000 h of data. Some technical failures or human errors resulted in a data loss of approximately 4%. After data cleaning (excluding measurements with high attenuation or error signal), 17% of all data was not considered for further analysis. This is a rather high number, but it was necessary to maintain data integrity and, because of the very large dataset, a conservative limit could be used setting a high standard for the data analysis.

All volunteers were nonsmokers and not exposed to secondhand smoke at home. Everyone was of working age and there was a small bias toward higher education. Most participants worked in an office, and everyone worked in an indoor environment. All 62 participants had a driving license, but not all couples owned a car. Participants were living in Flanders, Belgium (appendix FIGURE A4). An overview of personal and household characteristics is given in TABLE 6 and car attributes are summarized in the appendix TABLE A3.

TABLE 6: Characteristics of the study participants

		Summer	Winter
Personal characteristics			
Gender *	Male	8	23
	Female	8	23
Year of birth *	1951-1960	2	6
	1961-1970	7	12
	1971-1980	5	14
	1981-1990	2	14
Education / Highest degree *	Primary or secondary school	2	3
	Higher education, non-university	6	10
	Higher education, university	8	33
Working status *	Full-time worker	8	32
	Part-time worker	3	8
	Non-worker	5	6
Household characteristics			
Average household size *		3.88	3.65
Average number of cars per household *		1.38	1.48

* Results based on questionnaires filled in by the participants

Based on the activity diaries, it was calculated that volunteers spend 6.3% of their time (90 minutes per day) in transport; 35.5% of the day is spent sleeping (TABLE 7). The majority of trips were by car; but one third of all travel time was by active modes (bike, on foot). There are relatively more trips as car passenger in the weekend and on off-peak hours compared to car drivers. Train and metro are generally used by commuters, with a large share of trips in traffic peak hours and on weekdays. Trips as car driver, cyclist, bus passenger or walking are spread in the same way throughout the day and throughout the week. All light rail and metro trips are in urban areas, whereas car trips are often on highways (>25% of total time) and on rural roads (>30% of total time). More than 70% of the time, trips by bike or on foot are on urban or suburban roads.

TABLE 7: Time-activity pattern, contribution of each activity to total BC exposure, and contribution to inhaled dose (average of 62 participants)

Activity type	Time-activity pattern	Contribution to exposure	Contribution to dose
Home-based activities	29.9%	26.7%	21.8%
Sleep	35.5%	25.0%	13.9%
Work	17.0%	12.2%	12.8%
Social and leisure	6.3%	8.9%	12.8%
Shopping	1.1%	2.0%	3.2%
Other	3.9%	4.3%	5.6%
In transport	6.3% (100%)	21.0% (100%)	29.8% (100%)
Car driver	2.9% (45.3%)	12.3% (58.6%)	10.5% (35.2%)
Car passenger	0.7% (11.4%)	2.2% (10.3%)	1.7% (5.8%)
Bike	1.0% (15.7%)	2.5% (11.8%)	9.1% (30.5%)
On foot	1.0% (16.4%)	2.2% (10.5%)	6.6% (22.3%)
Train	0.5% (7.9%)	0.9% (4.2%)	0.9% (3.0%)
Light rail / metro	0.1% (0.8%)	0.2% (1.0%)	0.2% (0.7%)
Bus	0.2% (2.4%)	0.7% (3.6%)	0.7% (2.5%)

3.2.3.2 Personal exposure measurements

Average personal exposure was 1592 ng/m³, with a standard deviation of 468 ng/m³. This is comparable with the average concentration measured by the fixed suburban monitor (1620 ng/m³). The volunteer with the lowest personal exposure was exposed to 652 ng/m³ on average and the highest exposed participant to the study had a personal exposure of 2773 ng/m³. BC concentrations are lognormally distributed within each participant, meaning that there are a lot of 5-min observations with relatively low concentrations, and some observations where participants are highly exposed.

Lowest average concentrations were observed during home-based activities (1360 ng/m³), working (1077 ng/m³) and sleeping (1090 ng/m³). The highest average concentrations by far, were encountered while in transport (5132 ng/m³). High peaks are especially prominent in transport: 95th percentile is 15,569 ng/m³. Elevated average concentrations of 2445 ng/m³ and 2540 ng/m³ are observed for social and leisure activities, and for shopping respectively. Looking at microenvironments instead of activities, shows the same trends: high exposure in transport, lowest concentrations in private homes (1255 ng/m^3) and at work locations (1068 ng/m^3). Concentrations in transport were 2 to 5 times higher compared to concentrations encountered at home (appendix FIGURE A5 and FIGURE A6). Although the amount of time spent in transport is relatively small, this nevertheless corresponds to 21% of personal exposure to BC due merely to the high concentrations measured in transport (TABLE 7).

The transport category can be subdivided in different classes according to transport mode (FIGURE 18). Lowest concentrations were measured in trains, with a mean of 2394 ng/m³. Volunteers traveling with active modes, by bike or on foot, were confronted with higher average exposures ranging from 3175 ng/m³ to 3555 ng/m³. It should be noted that the average exposure of cyclists and pedestrians was 62% lower when the trip was a leisure trip, indicating the use of alternative routes instead of using the shortest (not seldom the most polluted) route as is more often the case for commute trips (see chapter 3.1). The exposure of volunteers traveling by motorized transport was highest (car driver: 6432 ng/m³; car passenger: 5583 ng/m³; bus passenger: 6575 ng/m³; and light rail / metro passenger: 5066 ng/m³). The results of the light rail / metro category are indicative since they only encompass 23 trips, although spread over different cities and weeks. In summary, the exposure-ratios of BC in different transport modes, in typical Belgian conditions, are: automobile:bicycle ratio = 1.77; automobile:foot ratio = 2; automobile:bus ratio = 0.96; and automobile:train ratio = 2.63.

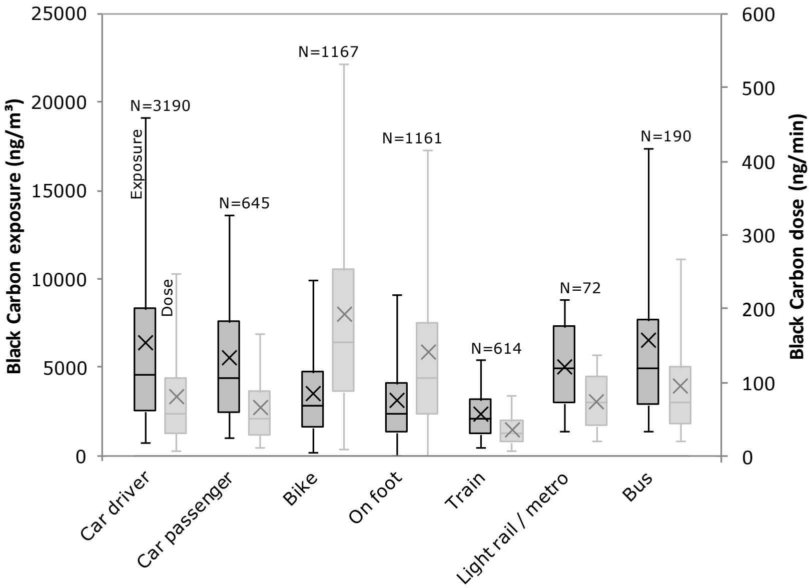


FIGURE 18: Personal exposure (black box plot) and dose (gray box plot) in different transport modes. The 'N=' indicates the number of 5-minute observations used to calculate the average and the percentiles. Represented are P5, 1st quartile, median, 3rd quartile and P95. The cross marks the average.

More than 65% of all trips were made either as car driver or as car passenger. An important determinant of the concentrations measured inside a car is the timing of a trip. FIGURE 19 shows the hourly variation in average concentrations: highest in-vehicle concentrations are observed during traffic peak hours (morning peak between 7 and 10 a.m., evening peak between 4 and 7 p.m.). This trend is less pronounced in other transport modes, probably because traffic congestion on rush hours affects car users more than e.g. cyclists or pedestrians. The impact of time-of-day on concentrations in microenvironments different from transport, is more limited although concentrations are still 33% higher in the evening (17-22 h) than during the day (8-16 h).

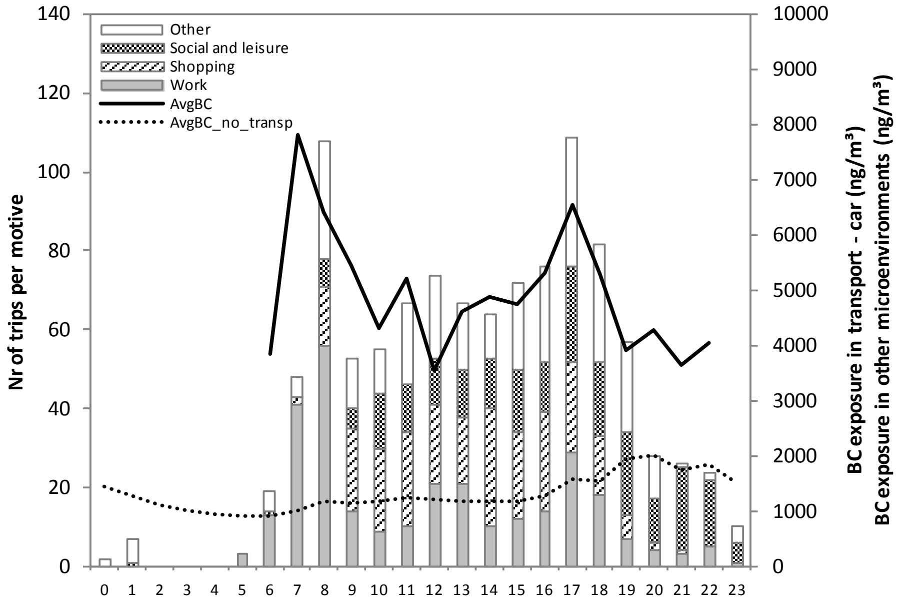


FIGURE 19: Timing of a car trip and the impact on in-car BC-concentrations. The black line shows the in-vehicle concentrations for every hour of the day. Each trip is assigned to the start hour of the trip. If less than 10 trips are available, the average was not included. The dashed line represents the average BC concentrations in all other microenvironments, excluding transport. The bars indicate the number of car trips and the corresponding trip motives on which the average concentration is based.

Day of the week affects personal exposure encountered in a car. We corrected the personal measurements for daily differences in background concentrations, but still differences between days appeared (FIGURE 20). Between working days, there are only minor differences in in-vehicle concentrations (from 5366 ng/m³ on Wednesday, to 5893 ng/m³ on Thursday), but they are in line with the traffic intensity on these days. Concentrations are lower on Saturdays (4459 ng/m³) and Sundays (3830 ng/m³). This difference between in-car concentrations on weekdays and weekend days was found to be statistically significant. A paired test was used to limit the influence of specific characteristics of each car: we assume that individuals drive the same car on all days of the week.

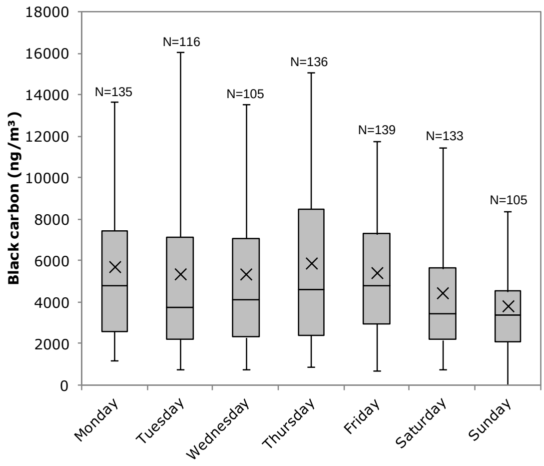


FIGURE 20: Timing of a car trip (day of the week) and the impact on in-car BC-concentrations. The 'N=' indicates the number of trips used to calculate the average and the percentiles. Represented are P5, 1st quartile, median, 3rd quartile and P95. The cross marks the mean value. In-car concentrations are significantly lower in the weekend than on weekdays (Paired T-test $p<0.01$).

3.2.3.3 Inhaled dose

Inhaled dose of the participant with the lowest average exposure to BC was 14,134 ng/day. The highest exposed individual inhaled on average 77,698 ng/day. In our study, the lowest and highest exposed individual also had the lowest and highest BC intake (dose), but this is not necessarily the case and depends on the executed activities.

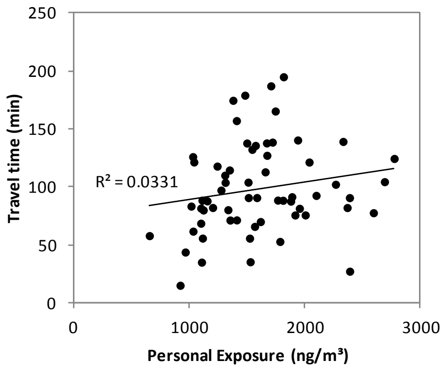
When comparing exposure and inhaled dose in FIGURE 18, it immediately shows up that the active modes contribute more to inhaled dose than to exposure. The highest dose is encountered on a bike, with an average dose of almost 200 ng/min. For inhaled dose, the ratios between different transport modes become: automobile:bicycle ratio = 0.41; automobile:foot ratio = 0.56; automobile:bus ratio = 0.82; and automobile:train ratio = 2.16.

On average the relative importance of the transport activity increases, up to 30%, when incorporating inhalation rates (TABLE 7). This difference is due to lower respiration during sleeping and higher respiration during active travel (e.g.

on a bike minute ventilation is 6 to 7 times higher compared to ventilation during sleep). The average daily dose incurred during transport is therefore similar to the average dose incurred from home-based activities, and much larger than the inhaled dose during the sleep activity.

3.2.3.4 Approximations for exposure and dose

Because multiple factors influence exposure in transport, it is not straightforward to relate a simple metric such as travel time to integrated personal exposure or inhaled dose (FIGURE 21). We demonstrated that average exposure in transport is very high, but also highly variable between individuals depending on the transport modes used, the timing of trips (time-of-day, day of the week), and possibly the geographical location of the trip (chapter 3.3). Limiting travel time to travel time by car reveals an equally poor correlation with personal exposure (appendix FIGURE A7). On the other hand, our results show a better correlation between average exposure in transport and average personal exposure (appendix FIGURE A8). Because we know that trips are responsible for 30% of daily inhaled dose, the relationship between travel time and dose is expected to be somewhat better: with 6% of explained variance, travel time is not very predictive for inhaled dose either. For comparison, the correlation between residential outdoor concentrations and personal exposure is 0.32 (appendix FIGURE A9).



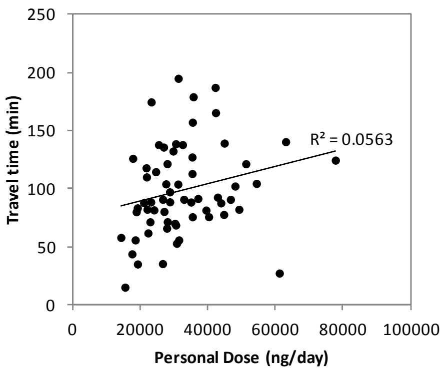


FIGURE 21: Correlation between personal exposure, daily dose and travel time. Each mark represents one of 62 volunteers.

3.2.4 DISCUSSION

In chapter 3.1 we already indicated that transport is responsible for almost a quarter of accumulated exposure, although individuals travel no more than 6% of the day. The results of the present more elaborate study confirm that transport indeed accounts for 21% of personal exposure. Our results further show that transport contributes up to 30% to the inhaled dose. In spite of the limited time spent in transport, more BC particles are inhaled during, on average, 90 minutes of transport compared to e.g. >500 minutes of sleeping or even during all other home-based activities combined. Nevertheless many epidemiological and HIA studies only include residential exposure and ignore exposure in transport; just recently several studies started to assess the health effects of in-traffic exposure to air pollution (McCreanor et al., 2007; Strak et al., 2010; Zuurbier et al., 2011).

The levels of exposure in transport depend on several factors; some of which were studied in depth in this paper: transport mode choice and timing of a trip. The most obvious factor influencing exposure during travel is transport mode choice. The highest average concentrations are encountered in motorized transport: car and bus. This is in line with findings of Adams et al. (2002) measuring Elemental Carbon in different transport modes, and with the review paper of Kaur et al. (2007) stressing the general trend in multi-mode exposure assessments on fine particulate matter: pedestrians and cyclists experience lower exposure concentrations than individuals inside vehicles. Zuurbier et al. (2010) measured soot in three different transport modes: car, bus and bicycle. Soot levels were highest in cars and buses, and lowest by bike along a low-traffic street. A review on ultrafine particle (UFP) exposure in the transport microenvironment, encompassing approximately 3000 individual trips in total, but originating from different studies, found overall mean UFP concentrations of 3.4; 4.2; 4.5; 4.7; 4.9 and 5.7 x 10^4 particles/cm^3 for the bicycle, bus, car, rail, walking and ferry modes, respectively (Knibbs et al., 2011). Boogaard et al. (2009) and Int Panis et al. (2010) reported smaller and inconsistent car/bicycle ratios for UFP in different towns. Particle number counts (PNC) were 5% higher on average in cars than on bicycles but this hides important local differences in either direction. In-vehicle BC concentrations were measured during 29 car trips by Rodes et al. (1998). Concentrations ranged from below the detection limit to 23,000 ng/m³; with an average of 6000 ng/m³ in urban Los Angeles, and 4000 ng/m³ as the statewide in-car average (Fruin et al., 2004). Sabin et al. (Sabin et al., 2005a; Sabin et al., 2005b) measured BC levels inside school buses between 900 ng/m³ and 19,000 ng/m³; this is comparable to concentrations measured in this study although our measurements were not limited to school buses or school bus hours. Bizjak and Tursic (1998) measured BC in buses as well and found higher concentrations in diesel buses (10,000-50,000 ng/m³), and lower concentrations inside a new gas-powered bus (5000-15,000 ng/m³). The portable aethalometer model AE51 was used by Weichenthal et al. (2011) to measure concentrations on a bike. Two types of routes were cycled: a high-traffic route (mean = 2520 ng/m³ (range 890-5670 ng/m³)) and a low-traffic route (mean = 1079 ng/m³ (range 173-3197 ng/m³)). These concentrations are lower than our mean concentrations for cyclists, but the trips in the study of Weichenthal et al. all took place outside peak hours (between approximately 11.30 a.m. and 12.30 p.m.), only in the warmer season and in an area with lower background concentrations. Apte et al. (2011) used the aethalometer model AE51 to determine BC exposure in auto-rickshaws in New Delhi, India. The geometric mean was 42,000 ng/m³, which is several times higher than BC concentrations observed in high-income countries. No studies were identified measuring exposure to BC in multiple modes, although the last study mentioned some limited in-car measurements. Because in FIGURE 18 the results were corrected for changing background concentrations, a direct comparison of absolute concentrations might be difficult (uncorrected concentrations are presented in the appendix TABLE A1). The overall ratios between modes of transport, presented in the previous section, can be biased by the fact that some modes of transport might be used preferentially in conditions that also affect ambient BC concentrations, e.g. in specific locations, rural versus urban, workdays versus weekend days, time-of-day or weather conditions. In the appendix TABLE A4 and A5 exposure ratios are split up for morning rush hours, evening rush hours and non-rush hours. The ratio automobile:(active modes) is lower on non-rush hours which probably reflects the fact that traffic congestion affects car users more than cyclists and pedestrians, as they are directly exposed to exhaust from preceding cars.

Clear differences in exposure between different transport modes appeared in our measurement campaign, but the large variation in BC concentrations in each transport mode reveals that multiple factors affect these concentrations. The timing of a trip seemed an evident, but little explored, parameter influencing in-transit concentrations. We saw elevated in-car concentrations on traffic peak hours, compared to off-peak hours; and elevated levels on working days compared to the weekend. Apte et al. (2011) looked at day of week differences in BC exposure; in line with our results, they found no consistent differences between working days. Sampling was limited to Monday through Friday, so any weekend effects could not have been detected. They did find time-of-day trends for BC: concentrations were 32% lower during the morning commute as compared to the evening commute, mainly attributable to the on-road environment. Hertel et al. (2008) looked at the impact of time-of-day on NOx concentrations on bike and bus. Higher than average concentrations were observed during morning rush hour compared to off-peak hours. The evening rush hour peak was less pronounced, probably because the period with high traffic intensities was also spread over more hours. The time-of-day variation in concentrations was mainly observed in buses on highly trafficked streets; the trend was less noticeable for bike trips on quieter streets. From Fruin et al. (2004) we know in-vehicle BC concentrations are highest when directly following diesel-powered vehicles. Considering the large number of diesel vehicles on Belgian roads (over 60% of all private cars in Belgium are diesel (NIS, 2010), a number that is remarkably higher than in neighboring countries), there is a high chance that participants drove behind diesel vehicles during rush hour. According to Westerdahl et al. (2005), on road BC concentrations also appeared to increase sharply as diesel truck traffic increased. A high proportion of traffic in Belgium is transit diesel truck traffic, especially on highways.

Trip motive can be defined as the activity that is performed on the destination side of a trip unless this is a 'home-based activity': in that case the activity at the origin side is defined as the trip motive (Cools et al., 2011). From FIGURE 19 it became clear that there are many work trips in the morning peak hours and there is a large share of leisure trips in the evening. Fewer trips have motive 'work' in the evening peak hour because of trip chaining. If we combine previous findings, it is straightforward to conclude that average in-car exposure is highest for trips with motive 'Work' (appendix FIGURE A10): these trips are often in traffic peak hours and on weekdays; and we know that during these time-periods the in-car concentrations are highest. Leisure trips are mainly in the evening or in weekends, and thus lower average exposure can be expected.

It is dose that is directly linked to health endpoints and thus relevant in epidemiological studies. Nevertheless many of these studies derive exposure and exposure-response functions based on the residential location only. In our study, we found that participation in traffic, especially active transport as a cyclist or pedestrian, increases inhaled dose more than proportionally. On average inhaled dose is clearly larger for active modes compared to all other modes, but on an individual basis the height of the dose depends on several factors. As already stated, it is very important to take timing and location into account. Breathing rates can also differ by individual and by the degree of physical exertion. Assumptions on minute volume were made based on literature and are an approximation of real minute volume. The question remains whether short periods exposed to elevated concentrations have any significant health effect on an individual. Weather (rain or heavy wind) might be a factor that shifts cyclists to days with higher BC concentrations. On the other hand, cyclists themselves try to avoid busy traffic or hilly terrain, take the least polluted and thus 'healthiest' route, and that way they decrease their exposure to traffic-related air pollution (Hertel et al., 2008; Int Panis et al., 2010; Zuurbier et al., 2010). Several studies already demonstrated that potential health effects caused by air pollution in traffic, are more than offset by the positive effects of active travel (de Hartog et al., 2010b; Rabl and de Nazelle, 2012; Woodcock et al., 2009).

Integrated weeklong personal exposure is highly variable between individuals, ranging from 652 ng/m^3 to 2773 ng/m^3. Those individual differences are much larger than population based estimates that take activities into account (Beckx et al., 2009b). Accurate exposure assessment, focusing on personal exposure rather than population based estimates, is thus critical to further reduce exposure misclassification in epidemiological studies. Personal measurements, as shown in this paper, are one way of estimating personal exposure more accurately. However, a methodology taking background variation into account is essential to compare measurements performed at different times. A combination of air quality models and activity-based models, producing time-activity schedules for every individual actor in a population, seem promising since models will be less hampered by sample size limitations than measurements. Using an activity-based model, exposures can be turned into inhaled doses in a straightforward way by assigning a minute ventilation to every activity.

It was impossible to relate in-vehicle concentrations to certain characteristics of the car since it is not known which trip is undertaken by exactly which car. This is a major drawback of this study, together with the lack of control over the routes taken, and the ventilation settings of the car. On the other hand we randomly selected different cars that were driven in a real-life setting, and from the questionnaires we had some basic information on the cars. This resulted in a very large and representative dataset, e.g. trips of different durations, trip chaining behavior was included, trips were geographically dispersed over a wider region, etc. The choice of volunteers participating in this study was somewhat biased, although a comparison of the activity diaries with a large cohort (over 1600 families) of the Flemish Travel Behavior Survey reveals good correspondence (Cools et al., 2011). Time in transport and the percentage of time in each transport mode, are also comparable to the Flemish average. We used only one monitor to correct for changing background concentrations; these measurements may not be completely representative for background concentrations in the entire study area. Unfortunately other fixed monitors, e.g. on rural background locations, were not available at the time of the measurements. Seasonal effects on BC exposure were not considered because we scaled the background-part of all observations based on concentrations measured at a fixed suburban background monitor. In contrast, we did observe a weekend – weekday effect because it was unrelated to changes in background concentrations.

In our study highest exposures were observed during travelling (especially car driving). Transport mode choice and timing of a trip proved important variables influencing exposure in transport. It was demonstrated that trips with motive ‘work’ had highest average in-car concentrations because they are mainly driven during peak hours. Inhaled dose per minute is highest during cycling and walking, but can be influenced by taking an appropriate route. We provided evidence that travel time is an unsatisfactory parameter to predict personal exposure to BC.

High peak hour concentrations may, at least partially, be caused by more frequent use of highways or urban roads (Sabin et al., 2005a). Westerdahl et al. (2005) suggested an impact of diesel truck volume on BC concentrations inside vehicles and the speed of a car may have an impact on the air exchange rates (Fruin et al., 2004), thereby influencing in-vehicle exposure to particles. Further research will therefore focus on these aspects as well as on the geographical location and road characteristics of the trips.

3.3 STREET CHARACTERISTICS AND TRAFFIC FACTORS DETERMINING ROAD USERS' EXPOSURE TO BLACK CARBON

This chapter is based on:

Dons, E., Temmerman, P., Van Poppel, M., Bellemans, T., Wets, G., Int Panis, L., 2013. Street characteristics and traffic factors determining road users' exposure to black carbon. Science of the Total Environment 447, 72-79.

3.3.1 INTRODUCTION

Black carbon (BC) is a component of both fine and coarse particulate matter (PM), though because of its small size, it is most strongly associated with the fine particle ($PM_{2.5}$) fraction (Smith et al., 2009; Viidanoja et al., 2002). Incomplete combustion of wood and diesel engine exhaust are the major environmental sources of BC pollution, respectively in rural and urban areas. Health effects of $PM_{2.5}$ are well documented (Brook et al., 2010) and BC is suspected to be one of the most harmful fractions responsible for health effects in exposed individuals (U.S.EPA, 2012b) and is a suitable indicator for assessing the health risks of traffic related air pollution (Janssen et al., 2011). Health outcomes associated with BC include cardiovascular effects (Adar et al., 2007; McCracken et al., 2010; Wellenius et al., 2012), respiratory effects (Lin et al., 2011; Patel et al., 2010) and mortality (Gan et al., 2011). Some of these effects can be demonstrated at ambient concentrations below 1 µg/m³ (Adar et al., 2007; McCracken et al., 2010). In addition BC contributes to global warming and has therefore gained more attention in recent years, resulting in its inclusion in several high level policy documents (UNECE, 2010; UNEP, 2011; WHO, 2012). BC is not yet regulated, but many $PM_{2.5}$ reduction measures, especially in the transportation field, should lead to reductions in BC exposure as well.

Although short in duration, exposure during transport can be important in integrated daily exposure to BC. In a personal exposure study in Belgium covering different degrees of urbanization and with a fleet dominated by diesel cars, concentrations of BC in transport have been shown to be a factor 2 to 5 higher compared to concentrations at home (see chapter 3.1 and 3.2). People spend on average 6% of their time in transport, but this leads to over 20% of daily integrated exposure, and up to 30% of the inhaled dose when taking breathing rates into account (see chapter 3.2). Travel behavior (Dhondt et al., 2012b), exposure in transport (Dons et al., 2012) and factors influencing in-vehicle air pollution (Fruin et al., 2004), are thus relevant factors to consider in integrated exposure assessment.

The mass of BC particles is measured in some stations of the national air quality network using an optical technique: MAAP (Multi-Angle Absorption Photometer). This technique is based on the measurement of the absorption and scattering of particles collected on a filter tape (Petzold et al., 2005); it thus relies on the optical properties of particulate matter. Atmospheric elemental carbon (EC) is a product of incomplete combustion. The terms EC and BC are often used interchangeably; however they are defined by the different measurement method applied (Quincey et al., 2009). EC is measured based on its chemical stability using thermal techniques. Fixed measurement stations are required to check compliance with national or international legislation. Many epidemiological studies use fixed site measurements as a surrogate measure of exposure. Unfortunately, fixed stations are a poor marker for personal exposure, especially for pollutants highly variable in time and space like BC (Koutrakis et al., 2005).

According to Ott (1982) a person i is exposed to concentration c of a pollutant at a particular instant of time when *person i comes into contact with the pollutant at concentration c*. This definition can be decomposed into two events occurring at the same time: *person i is present at location x,y,z at time t*, and *concentration c is present at location x,y,z at time t*. Whether an individual is highly exposed, thus depends on the concentration levels the person encounters in the microenvironments visited over a day. Improving exposure estimates has been recognized as an important topic (Int Panis, 2010) and the US EPA has launched several programs to achieve better indoor (SHEDS) and personal (EMI) exposure estimates (Breen et al., 2010). For static microenvironments ('places'), it is fairly straightforward to model outdoor concentrations by using an air quality model, and if desirable supplemented with an indoor air model (Burke et al., 2001). For mobile environments ('in transport') estimating concentrations is much more difficult because the location and conditions are constantly changing. One possible approach is to calculate concentrations on center points of road segments using a land use regression or dispersion model, and to intersect this polyline map of pollution with trips (Mölter et al., 2012). A similar approach uses a concentration grid, e.g. as an output of a dispersion model, and time-weights this grid with trips crossing different grid cells (Marshall et al., 2006). Other studies either ignore exposure in transport (Hatzopoulou and Miller, 2010), or use concentrations modeled (Dhondt et al., 2012b) or measured (Beckx et al., 2009b) at fixed stations near busy roads. Kaur et al. (2007) stated that four factors can contribute to BC levels in transport: personal factors, mode of transport factors, traffic factors and meteorological factors. The impact of personal factors (time-activity pattern, timing of trips and breathing rates) and transport mode factors are discussed in chapter 3.2. The aim of the current analysis is to determine traffic factors influencing exposure to BC in transport and to derive an 'in-transport exposure model'. The analysis is based on measurements of BC in Flanders, Belgium. Road and traffic data associated with a road network were linked in a GIS with geocoded BC-measurements. Relevant factors are identified and discussed. A simple set of models is constructed to estimate average BC exposure during trips based on data that is readily available from most traffic models.

3.3.2 MATERIALS AND METHODS

A set of mobile measurements was collected between April 2010 and July 2010, and between December 2010 and February 2011. Sixty-two volunteers measured their personal exposure to BC continuously for 7 days, while also logging GPS positions and reporting detailed time-activity patterns. All participants were living in Flanders, an urbanized region in the north of Belgium (13,521 km²; 6,251,983 inhabitants). A description of the study set-up is given below; technical specifications of the portable devices used are summarized in TABLE 8.

TABLE 8: Technical specifications of the portable devices

	Time base	Tech Specs
GPS receiver	1-s	GPS receiver integrated in PDA (Personal digital assistant, type MIO 168, weight 147 g, 4 hour run time on single battery charge if in use, 12 GPS channels)
Electronic diary	5-min	Custom designed software installed on PDA
Micro-aethalometer AE51	5-min	Flow rate 100 ml/min, weight 280 g, 24 hour run time on single battery charge, optical measurement of BC

3.3.2.1 Time-activity diary and GPS logging

A handheld computer or PDA (Personal Digital Assistant) was equipped with the software tool PARROTS (PDA system for Activity Registration and Recording of Travel Scheduling) to facilitate the registration of activities and locations visited (Kochan et al., 2010). The GUI of PARROTS was developed as a continuous timeline, and participants had to build and annotate time blocks in a visually appealing way by using drop-down menus, check boxes, etc. The data provided had to be accurate to within 5 minutes. Start time and end time of each activity had to be indicated, together with the type of activity choosing from 13 predefined activity categories. PARROTS offers a choice of several transport modes: car driver, car passenger, motorcycle, bike, on foot, bus, train, light rail / metro, taxi, or 'other'. Trips during which participants used multiple modes had to be reported as one trip, but each part of the trip had to be detailed (transport mode, travel time, waiting time). During the week, every diary was repeatedly checked on consistency and completeness: no gaps or overlaps were allowed. Afterwards the time-activity data was automatically processed in SAS 9.2.

A GPS receiver was integrated in the PDA, logging positions on a one second time base. Participants had to initiate the GPS each time they started a trip, and stop it when the trip was finished to prevent unnecessary logging and to conserve the battery. Sporadically volunteers forgot to start or stop the GPS, resulting in loss of information on certain trips. Since the accuracy of the GPS signal is influenced by the number of satellites in view, GPS waypoints were cleaned such that each observation was made with at least 5 satellites: this was the case for 88.6% of all observations in transport. Rail-based modes are not explored further because of regular failure of reception of satellite signals (also observed by Bohte and Maat (2009) and Beekhuizen et al. (2013)). If GPS coordinates indicated a trip, but the diary reported an activity on a fixed location, the observation was omitted because the lack of information on e.g. transport mode.

3.3.2.2 BC exposure monitoring

A portable aethalometer, MicroAeth Model AE51, measured BC on a 5-min time resolution (AethLabs, 2011). This aethalometer can monitor with a higher temporal resolution, up to 1 second, but a 5 minute time base was used to improve precision and to extend battery life and run time. A short tube was attached to the inlet of the aethalometer giving volunteers the possibility to put the aethalometer in a purse or backpack while still measuring ambient air. The teflon-coated borosilicate glass fiber filter was replaced every two days by the participants to prevent filter saturation. Clear instructions were provided to do this correctly. Volunteers were instructed to take the aethalometers wherever they went, although indoors it was allowed to keep it static in a room where the majority of the time was spent (e.g. in the living room, in the office). Despite the obtrusive nature of personal exposure monitoring, this study tried to be as least demanding as possible and participants were encouraged to follow their daily habits and execute their regular activities.

Before analyzing the BC data, several data cleaning steps were necessary. BC measurements with high attenuation (ATN > 75) and measurements showing an error code were excluded, as described in chapter 3.2. An intercomparison of the thirteen micro-aethalometers resulted in correction factors for device specific deviations in the range of 1 to 23%. Before each measurement campaign, the air flow of the aethalometers was checked and fixed at 100 ml/min. Because not all measurements were made simultaneously (16 weeks, 112 different days), a daily rescaling was applied to account for changing background concentrations. More details on the methodology can be found in chapter 3.2.

3.3.2.3 Data integration

Individual waypoints were combined with data from the electronic diary and with BC concentration information. Travel speed was automatically calculated based on the GPS data. Due to the different temporal resolution of the datasets, up to 300 waypoints are attributed to the same BC concentration (FIGURE 22).

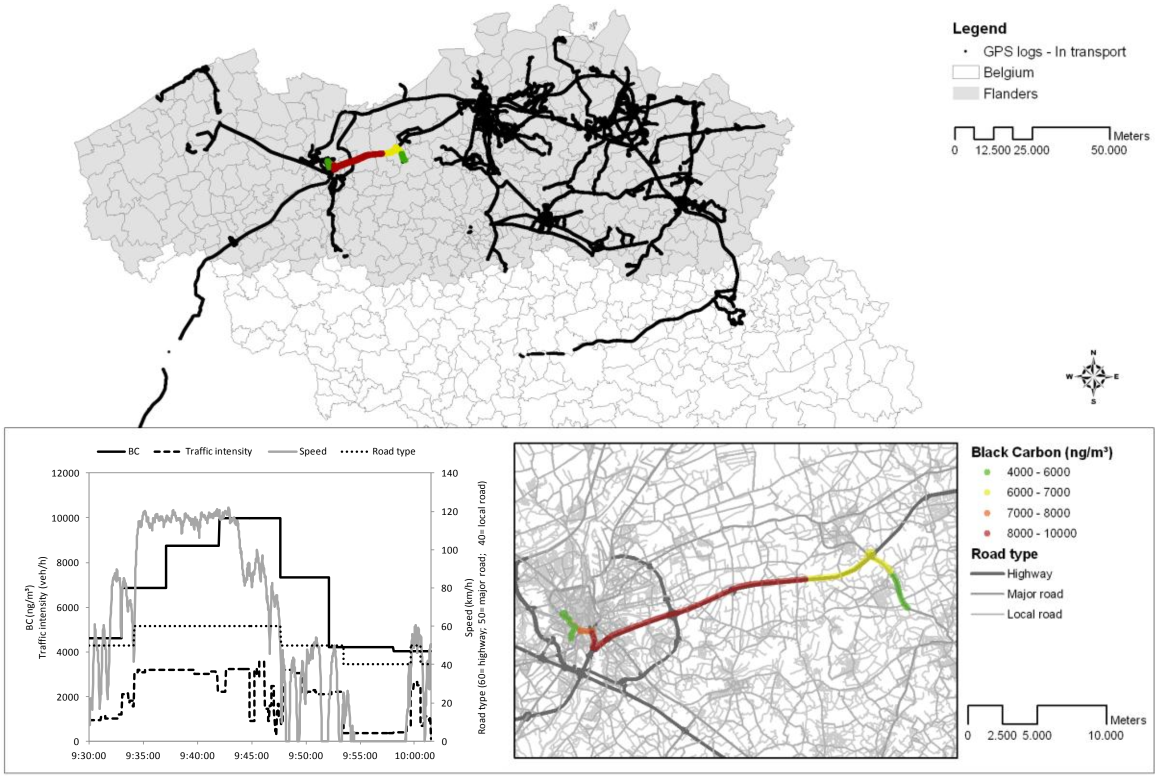


FIGURE 22: Top: Overview of all GPS logs captured by 62 volunteers. Bottom: excerpt of a single car trip illustrating the different temporal resolution between GPS waypoints and BC measurements (trip starts on a major road, continues on a highway and ends on local roads).

In ArcGIS 10 waypoints were joined to the nearest road (details of the road network are given in the appendix TABLE A6 and FIGURE A11). The average distance to the nearest road gradually decreased when the number of satellites increased to level off from 8 satellites onward; waypoints based on fewer than 5 satellites were excluded (appendix FIGURE A12). If the distance to the nearest road was larger than 30 m, no road characteristics were linked to the BC-observation assuming that this was not the road where participants were traveling on. The 30 m cut off was decided arbitrarily, but based on decay rates for BC near roads (Beckerman et al., 2008; Karner et al., 2010; Zhu et al., 2002) and also taking into account the known inaccuracies in the road network (appendix FIGURE A13). Trips outside of the study area, in casu Flanders, were automatically excluded by using the above criterion. The conversion steps between the datasets are summarized in FIGURE 23.

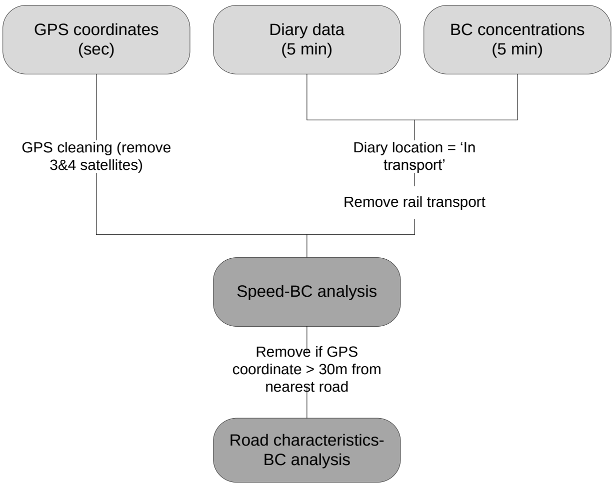


FIGURE 23: Schematic overview of available datasets, data cleaning, and data analysis

Annual average daily traffic, associated with every road link, was converted into instantaneous traffic intensities (using standard factors see appendix TABLE A7) taking into account the hour and day a person was present at a certain link. We thus introduced a link between traffic intensity and hour of the day.

Eight road types were identified and grouped in three broader categories: highways, major local roads (primary and secondary roads) and other local roads (based on road classification; appendix FIGURE A11). Additionally the variable 'urbanization' was used to assign every road to one of four categories: highway, urban, suburban or rural (appendix FIGURE A11). Traffic on every network link has a modeled speed; this speed slightly deviates from the maximum (free flow) speed. All map attributes were linked with BC concentrations measured in transport.

3.3.2.4 Statistical analysis

In the analysis, motorized modes (car, bus) and active modes (bike, on foot) are combined to ensure data representativeness; the groups were formed taking into account the position of each road user on the road, and the enclosed nature of vehicles. Descriptive statistics are calculated primarily; afterwards simple models are selected using the forward stepwise regression technique, with the adjusted R^2 as entry criterion. This procedure is very similar to the land use regression (LUR) technique frequently used to model air pollution at residential addresses, but it is now used with mobile measurements. A precondition for using regression models is the absence of autocorrelation in the dataset; this is examined by calculating the correlation between a measurement at time *t* and measurements at time *t-1*, *t-2*, etc. Model results are summarized in a tree using logical if-then conditions.

3.3.3 RESULTS

3.3.3.1 Dataset

Our dataset contains 7039 BC measurements (5-min average) when participants' diaries indicated they were 'in transport'. For approximately 70% of all motorized trips and 60% of all active trips registered in the diary, GPS logs were recorded. In total, 906,112 GPS waypoints (1-s) were registered while traveling with motorized transport (car or bus), and 385,453 waypoints were logged while traveling on foot or by bike (FIGURE 22). Main reasons for missing GPS data are not initiating the PDA or signal loss. This resulted in a dataset of 4462 geocoded 5-min BC-measurements. Overall, 18% of the original data were lost because BC measurements were missing; another 30% of the observations were lost because no GPS-data was available; because of the removal of points >30m of a road with known traffic intensities 33% of all GPS logs were not included in the analysis. Almost a quarter of the remaining observations in motorized modes was on highways, whereas 28% was on major roads and 48% on local roads; most trips with active modes were on local roads (88%). The range in BC exposure is larger for participants traveling by motorized transport (IQR=6930 ng/m³) compared to exposure of active travelers (IQR=3439 ng/m³).

3.3.3.2 Timing of a trip

During traffic peak hours (morning peak between 7 and 10 a.m., evening peak between 4 and 7 p.m.) exposure of motorists is higher than during off-peak hours (Peak: 9708 ng/m³ - Off-peak: 6957 ng/m³ - Weekend: 6078 ng/m³). The hour-to-hour variation in in-vehicle concentrations is presented in chapter 3.2. For the active modes a similar peak/off-peak pattern emerges (Peak: 4827 ng/m³ - Off-peak: 4310 ng/m³ - Weekend: 3353 ng/m³), however the difference between peak and off-peak is smaller.

3.3.3.3 Travel speed and traffic speed

FIGURE 24 shows the BC exposure related to transport mode, travel speed (GPS speed) and modeled road speed. No distinction is made for road types. We observed a nonlinear relationship between in-vehicle BC concentrations and GPS-based travel speed (FIGURE 24, left): there are peaks at approximately 20 km/h and 100 km/h and lower concentrations at speeds around 50-70 km/h for car drivers. Lower speeds are encountered in urban stop-and-go traffic.

Acceleration and proximity to other vehicles subsequently result in higher BC emission and higher exposure of following car drivers. Above 80 km/h, there is a steep increase. These speeds are typically linked to driving on primary roads and highways where traffic intensities are higher and a larger fraction of heavy traffic is present. At very high speeds, over 130 km/h BC exposure decreases again, probably because cars only get to this speed at off-peak hours when no other traffic is nearby. A similar pattern emerges for motorized traffic and (GIS-based) road speed, as vehicles often drive close to maximum speed (FIGURE 24, right).

For active modes, the relationship is clearly different for driving speed (the speed of the cyclist/pedestrian) and road speed (the speed of the motorized traffic). Driving speed (GPS speed) does not influence BC concentrations for the active modes. Cycling or walking on roads with low modeled car speeds, results in higher than average exposure as a result of higher emissions and closer proximity to motorized traffic. When the road speed increases, exposure of the cyclist or pedestrian decreases; this can be explained by lower emissions and the increasing lateral distance to the roadway. At higher road speeds (+60 km/h) an increase in exposure of active travelers is seen similar to the increase in exposure of motorists, indicative of increasing emissions although the uncertainty is larger because of the limited number of observations.

On highways, the relationship between speed and BC concentrations is different compared to the general speed-BC association presented above: highest exposures are observed at very low speeds and exposure continuously decreases from there. The average in-vehicle concentrations on highways decline from 15,517 ng/m^3 (speeds up to 20 km/h) to 9754 ng/m^3 at speeds over 120 km/h (appendix FIGURE A15). For major and local roads a similar BC increase at lower speeds is observed; although the absolute height of concentrations is lower (for major roads: 9768 ng/m^3 for speeds up to 20 km/h, to 7623 ng/m^3 at speeds over 80 km/h; for local roads: 7147 ng/m^3 for speeds up to 20 km/h, to 6034 ng/m^3 at speeds over 60 km/h). A combination of these three relationships results in the general BC-speed relationship (FIGURE 24), but taking into account frequency of use of different road types.

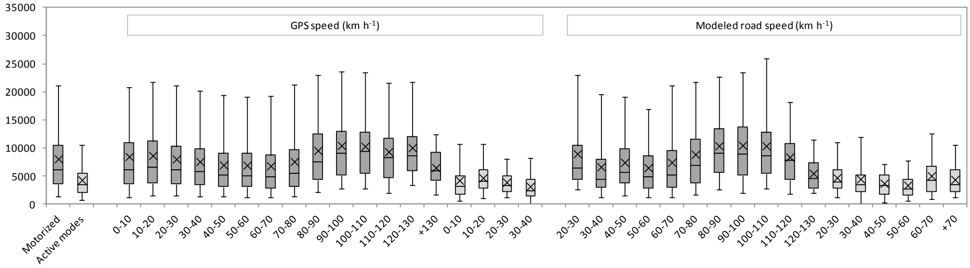


FIGURE 24: BC exposure (ng/m³) is presented on the y-axis and this is related to transport mode, travel speed, and modeled road speed on the x-axis. Motorized modes (car, bus) are shown in darker gray, active modes (bike, on foot) in lighter gray. Represented are P5, 1st quartile, median, 3rd quartile and P95. The cross marks the mean value.

3.3.3.4 Urbanization, road type, and traffic intensity

The degree of urbanization correlates with air pollution levels; it is thus expected that degree of urbanization also affects in-traffic BC exposure. Average in-vehicle exposure is highest in urban areas and lowest in rural areas (Urban: 9568 ng/m³ - Suburban: 7208 ng/m³ - Rural: 6105 ng/m³; figure 4). In this study, 24% of all observations were on highways, 15% on urban roads, 29% on suburban roads and 32% on rural roads. A similar trend in exposure can be observed for the active modes, although the absolute and relative differences are smaller. The largest fraction of active trips was observed in urban (38%) and suburban areas (40%).

On highways, exposure of motorists is higher compared to exposure on major roads, and almost double the in-vehicle concentrations measured on local roads (Highways: 10,680 ng/m³ - Major roads: 8359 ng/m³ - Local roads: 6501 ng/m³; FIGURE 25). For active modes the relationship is less intuitive: it appears that the exposure of cyclists and pedestrians is lower near major roads compared to local roads.

BC exposure also shows a clear increasing trend with traffic intensity: the more vehicles travel on the same road the higher the in-vehicle BC concentrations, only the highest category (+4000 veh/h) deviates slightly from this line. Large differences exist between quiet roads and busy roads: the average concentration in cars changes from 5646 ng/m³ to 13,623 ng/m³. For active modes a similar trend was observed on local roads although absolute concentrations are much lower (appendix FIGURE A16). On major roads exposure of cyclists/pedestrians is constant with increasing traffic intensities. This can probably be explained by the increasing lateral distance between the active travelers and the main roadway.

Traffic intensity is positively linearly related to in-vehicle BC exposure; moreover this relationship holds for every road type (appendix FIGURE A17). There are two exceptions: highway entries and exits have low traffic intensities but have an elevated in-vehicle concentration (due to heavy acceleration and proximity to the highway), and at very high traffic intensities above 4000 veh/h concentrations seem to level off (possibly influenced by the lower and less representative number of observations in this group).

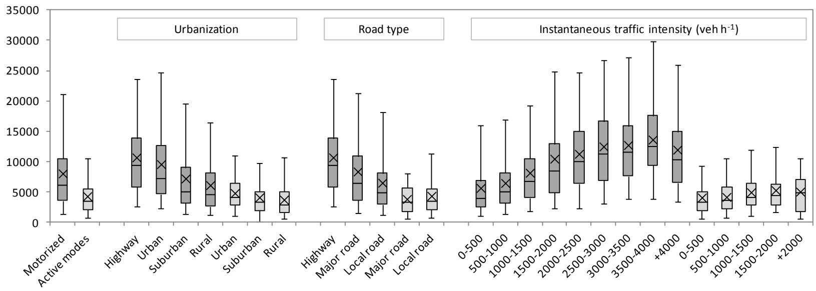


FIGURE 25: BC exposure (ng/m³) is presented on the y-axis and this is related to transport mode, urbanization, road type, and instantaneous traffic intensities on the x-axis. Motorized modes (car, bus) are shown in darker gray, active modes (bike, on foot) in lighter gray. Represented are P5, 1st quartile, median, 3rd quartile and P95. The cross marks the mean value.

3.3.3.5 Associations between variables

Because of the abrupt changes in exposures while in transport, sequential 5-min measurements were not autocorrelated (correlation with previous measurements dropped quickly). For motorized transport, instantaneous traffic intensity, timing (peak, off-peak, weekend; as indicator variables), and urbanization respectively, explain concentrations (model R^2=0.17) (appendix TABLE A8). If traffic intensities are unknown, timing and urbanization determine BC concentrations but the model loses explanatory power (R^2=0.12). Trip duration and road type (correlated with traffic intensity and urbanization, TABLE 9) don't contribute in a multiple regression model. Speed is offered to the model as a categorical variable but it was not included in the final model. The effect of speed on BC exposure is small in absolute numbers compared to the effect of some other variables; furthermore speed is highly correlated with road type. If urbanization type is unclear or unknown, trip duration (available from most traffic models) could be used as a proxy for the use of different degrees of urbanization (e.g. longer trips (>45min) are on highways for at least 45% of total time, whereas short trips (<30min) use highways only sporadically). Models perform worse for active modes when considering Pearson correlation: only timing and urbanization are associated with BC exposure (model R^2=0.02) (appendix TABLE A9). Results can be presented as a tree, enabling fast and easy to use reference values for exposure in transport (appendix FIGURE A18 and A19).

TABLE 9: Correlation matrix between BC levels and individual variables (Spearman's rank correlation coefficient) for motorized and active modes (italic) respectively.

Motorized _Active_	BC	Traffic intensity	Urbanization[a]	Road type[b]	Road speed	Travel speed	Time[c]
BC	1						
Traffic intensity	0.42* _0.17*_	1					
Urbanization[a]	-0.32* _-0.20*_	-0.45* _-0.25*_	1				
Road type[b]	-0.30* _0.02*_	-0.66* _-0.43*_	0.62* _0.06*_	1			
Road speed	0.20* _-0.03*_	0.50* _0.22*_	-0.27* _0.61*_	-0.74* _-0.37*_	1		
Travel speed	0.11* _0.09*_	0.31* _0.02*_	-0.30* _-0.06*_	-0.55* _0.003_	0.55* _0.04*_	1	
Time[c]	-0.27* _-0.21*_	-0.15* _-0.17*_	0.03* _0.03*_	0.08* _0.08*_	-0.08* _-0.07*_	-0.05* _-0.09*_	1

* significant at 1%.

[a] 4 classes: Highway, urban, suburban, rural

[b] 3 classes: Highway, major road, local road

[c] 3 classes: Peak, off-peak, weekend

3.3.4 DISCUSSION

Numerous variables influence personal exposure in transport. Kaur et al. (2007) classified potential confounders in four categories: personal factors, mode of transport factors, traffic factors and meteorological factors. Personal factors (e.g. breathing rates) and the impact of transport mode choice are discussed in chapter 3.2. There it was found that exposure in motorized transport is clearly higher than exposure while walking or biking (avg: 6323 ng/m³ versus 3365 ng/m³), although when accounting for inhaled doses, this relationship is reversed. The impact of timing of a trip on exposure was also explored. On traffic peak hours in-car BC concentrations are ~2000 ng/m³ higher than average. Differences in in-car concentrations between weekdays and weekend days are prominent, even when correcting for differences in background concentrations; this temporal trend is in accordance with concentrations measured on fixed locations near highways in Flanders (Van Poppel et al., 2012b). The focus in this paper is on traffic factors, like traffic intensity or road type, and their impact on personal exposure in transport microenvironments. Meteorology is not considered since all observations are adjusted for changes in background BC concentrations, which includes corrections for most meteorology effects except for the very local.

Travel speed is related to BC exposure: for motorized transport in-vehicle concentrations are elevated at lower speeds (up to 30 km/h) and at speeds above 80 km/h. Using road speed instead of travel speed does not influence our results significantly. The speed-exposure function shows a similar pattern than the BC emission function with a trough at speeds around 70 km/h, and with a sharp increase at higher speeds (Kristensson et al., 2004). At lower speeds in urban traffic, or when congestion is present on highways, following distance to other vehicles decreases and emissions can infiltrate in vehicles nearby. This results in higher BC exposure, since Fruin et al. (2004) concluded that the main source of BC in a car, are the emissions of the car followed. For active travelers, travel speed and BC exposure are unrelated (speed dependent breathing rates affect dose not exposure); the speed of motorized traffic on the other hand does seem to influence exposure of cyclists and pedestrians. In-vehicle exposure is highest while driving on highways and lowest on local roads. For cyclists and pedestrians, exposure on local roads tends to be higher compared to exposure when moving near major roads. In Flanders, cyclists on local roads are often mixed with motorized traffic and thus closer to the most important source of BC, while on major roads ‘cycle tracks’ are used to physically separate cyclists from faster (motor vehicle) traffic, resulting in lower exposures (de Hartog et al., 2010b; Int Panis et al., 2010; Thai et al., 2008). Cycle tracks are always located within 30 m of the main road to which the concentration was attributed. Dedicated off-road bike paths without any motorized traffic, or small local roads are not included in the digital road network, and thus not included in the dataset studied here. However it is expected that exposure will be at background levels on these roads. Concentrations presented here for local roads, either urban or rural, are therefore slightly overestimated (appendix FIGURE A14). In urban areas exposure is higher compared to more rural areas, this holds for both motorists and for cyclists and pedestrians. In-traffic exposure increases with traffic intensities for all road users, although this increase is much steeper for motorists. In several studies it is shown that higher traffic intensities lead to an increase in concentrations inside vehicles, particularly because of the increase in the number of pollution sources and a reduction in the distances between the vehicles (Chan and Chung, 2003; Zagury et al., 2000). A deviation from the linear function between traffic flow and BC exposure, are highway entries and exits: they are categorized as ‘highway’ on the digital map, have low traffic intensities, but are nevertheless associated with high BC concentrations. Probable causes are the extra emissions during acceleration and the presence of a nearby highway with heavy traffic flows. The traffic flow – BC exposure function seems to flatten out on highways with very high traffic intensities at concentration levels of ~13,000 ng/m³, but this might be influenced by the lower number of observations at high traffic intensities. Overall, concentrations are always higher for motorists than for active travelers on roads with similar characteristics.

Traffic intensity and timing of a trip are the main drivers for BC exposure during motorized transport. For active travelers, traffic intensity is not very predictive, probably caused by the greater distance of biking facilities to busy roads and they are thus further away from the most important source of BC. Urbanization proves to be predictive in both models, reflecting regional background concentrations on top of the local contribution caused by traffic. Road type is not included in the models, but this variable is highly correlated to both traffic intensity and urbanization (TABLE 9). R^2 values are low because of the broad range in measured concentrations within each category, but on the other hand the models are capable of predicting average concentrations with high precision. The LUR-like models can be applied to estimate exposure in transport at a certain point in time, but they can also be used to estimate exposure during a complete trip.

The results presented here summarize 'external' exposure of travelers and do not take into account inhaled doses: active travelers had a median exposure of 3547 ng/m³ and motorists had a median exposure of 6146 ng/m³. This is remarkably higher than the average in-home exposure for the same 62 volunteers (1255 ng/m³) (see chapter 3.2). Elevated exposure to BC comes at a cost: important health effects are associated with small increases in exposure, both over longer time periods and during short exposure peaks, e.g. in traffic. McCracken et al. (2010) found that an IQR increase in annual BC of 250 ng/m³ was associated with a 7.6% decrease (95% CI, –12.8 to –2.1%) in leukocyte telomere length, a marker for biological age and inversely associated with risk of cardiovascular disease (CVD). An IQR change in the 24-hour mean concentration (459 ng/m³) results in a 19% decrease (95% CI, -21 to -17%) in high frequency power when studying heart rate variability (Adar et al., 2007). BC averaged over 24 hours was strongly associated with exhaled nitric oxide, an acute respiratory inflammation biomarker: a 16.6% increase (95% CI, 14.1 to 19.2%) per IQR increase in BC (4000 ng/m³) (Lin et al., 2011). Patel et al. (2010) found that an increase in exposure to BC with 1200 ng/m³ led to significant acute respiratory effects in adolescents. Long term exposure to BC (IQR increase of 800 ng/m³) was associated with a 3% increase in coronary heart disease (CHD) hospitalization and a 6% increase in CHD mortality (Gan et al., 2011).

This study has some strengths and weaknesses. Performing an analysis with GPS-data, inherently leads to at least two sources of error: signal and map (Marchal et al., 2005). The accuracy of the GPS signal can be influenced by the number and the constellation of satellites, and the built environment (Kerr et al., 2011; Schuessler and Axhausen, 2009). When the GPS device is trying to get a fix, or physical structures block a clear view from the sky, e.g. in tunnels, a temporary interruption of the signal is probable. As pointed out by e.g. Morabia et al. (2009) and Beekhuizen et al. (2013), GPS signal errors in cities often are due to the urban canyon multipath-effect of tall buildings near the GPS receiver. If the number of visible satellites increases, e.g. more than 4 (cut-off value used in this study), the reliability of the signal improves. The inaccuracy of the road map is a second source of error. The digital road network regularly cuts off corners or oversimplifies neighborhoods with low traffic intensities; on most roads this error is limited to 30m. One of the consequences is that points are deleted because the nearest road is further than 30m, although the GPS position was correct. A potential improvement to the current data analysis is the use of more complex map matching algorithms instead of using the nearest road (Marchal et al., 2005). But although even if a person is not present on a road where he is mistakenly linked to, that person will be in close vicinity to this road and feel it's impact (e.g. on a bridge over a highway). Due to the 30m-restriction, only a small portion of our data will be linked to a wrong road segment, and the errors will be limited. The signal, map and map matching errors do not influence our overall conclusions but they may reduce the power to detect subtle effects or interactions among predictors. Most GPS-studies are constrained by the use of a GPS logging device only (Steinle et al., 2013). Single trajectories need to be analyzed and annotated using semantic trajectories data mining techniques, while in this study the transport mode and trip motive were reported by the volunteers (Bohte and Maat, 2009; Schuessler and Axhausen, 2009; Wu et al., 2011a). Modes were afterwards aggregated in two groups (i.e., car/bus, walk/bike) because average concentrations were similar and because of the common enclosure in a vehicle cabin; when taking into account breathing rates a disaggregated approach is advisable.

In our survey setup we gave priority to battery lifetime of the micro-aethalometers by measuring on a lower time resolution, combined with an intermediate sampling rate for audible noise constraints. This resulted in a temporal resolution mismatch with the 1-second GPS data. As illustrated in FIGURE 22, immediate impacts on BC exposure, for example when changing from a local road to a highway, are blurred by the lower time resolution; therefore we needed more measurements to draw the same conclusions. A sensitivity analysis excluding BC measurements with less than 75% of GPS points on the same road type nevertheless showed very similar results (appendix FIGURE A20 and A21). Future research should focus on measuring air pollution with a higher temporal resolution to improve the spatial resolution, to narrow the concentration range in each category and to improve the model R^2.

Impacts of the road environment on in-vehicle concentrations are immediate. There is no build-up of particles in a vehicle because of the fast exchange between indoor and outdoor air (Fruin et al., 2004). Without the recirculation setting switched on, the air inside a vehicle is renewed 63 times/h, but this may depend on ventilation settings, vehicle type and travel speed (Hudda et al., 2012; Knibbs et al., 2009). People who are not enclosed in vehicles, but are walking and cycling experience a similar effect, making it beneficial for health to take a parallel but quieter route where the cycle track is preferably located away from traffic (Delgado-Saborit, 2012; Zuurbier et al., 2010).

In conclusion, there exists a positive association between in-vehicle BC exposure and traffic intensity. In urban areas exposure of motorists and active travelers is higher compared to exposure in more rural areas; the same holds for highways versus local roads for motorists. In-vehicle exposure is highest while driving with speeds around 20 km/h and 100 km/h. There is no build-up of particles inside vehicles, but the duration of a trip is linked to the time spent on each road type. Traveling in traffic peak hours, increases exposure of all road users. Simple linear regression models can be built to predict exposure in transport based on widely available data. Because of the high variability of BC concentrations and the 5-min temporal resolution of the measurements, such models have a relatively low predictive power for specific conditions (low R^2), but are nevertheless very good in predicting average concentrations. On the other hand these regression models may still drastically improve personal exposure estimates because the concentrations in transport are much higher than residential concentrations. In future work we plan to use these estimates in combination with a classic LUR model to make integrated estimates of personal exposure based on activity-based models.

4. MODELING PERSONAL EXPOSURE

4.1 URBAN AND REGIONAL LAND USE REGRESSION MODELS FOR AIR POLLUTION EXPOSURE ASSESSMENT

This chapter is partly based on work performed in:

Van Poppel, M., Dons, E., Peters, J., Brabers, R., Damen, E., Daems, J., Van Laer, J., Berghmans, P., Int Panis, L., 2012. Metingen van ultrafijn stof in Vlaanderen op hotspot(s) voor de blootstelling aan verkeerspolluenten. VITO, p. 220.

4.1.1 INTRODUCTION

Exposure of a population to air pollution can be modeled by combining two datasets: predicted concentrations and time-activity patterns of individuals (Beckx et al., 2009a; Briggs, 2005; Int Panis, 2010). The latter can be obtained through large scale and lengthy questionnaires studying the time use of individuals in a population. Based on these revealed diaries probabilities can be assigned to individual decisions while building a diary: an individual with certain characteristics is working or not, decision on work location, length of a shopping activity, whether a trip is necessary, transport mode choice, etc. These so called activity-based models thus predict diaries of virtual agents (Davidson et al., 2007; Kitamura et al., 2000). Nowadays GPS loggers are a less intrusive and relatively cheap way of effectively measuring revealed time-activity patterns and the activity space of real-life individuals (Wu et al., 2011a). The first dataset needed to estimate exposure, ambient air pollution, can be measured at selected locations, but in larger cohorts concentrations need to be modeled with geostatistical techniques or physico-chemical models. Dispersion models predict concentrations on a grid; in an urbanized region and for traffic-related pollutants, large grid cells (with sides of 100-3000m) are problematic because concentrations change very locally. In epidemiology, land use regression (LUR) models are often used to predict exposure on residential locations. This methodology is preferred over dispersion models because of its flexibility, good performance, and transparency. LUR models establish a link between air pollution measurements on fixed locations and geographical variables (Briggs et al., 1997; Hoek et al., 2008). Because continuous measurements of the official air quality network are not spatially dense enough, a purpose designed network is often set up to develop LUR models. Study areas range from smaller urban areas up to international LUR models (Beelen et al., 2009). In peer-reviewed literature, it is generally accepted that the transferability of LUR models between study areas is rather limited (Clougherty et al., 2008; Johnson et al., 2010; Smith et al., 2006). LUR models use area specific predictors, and they often differ from one region to another or between countries (e.g. distance to the shore, impact of altitude, percentage of diesel traffic in a country). Vienneau et al. (2010) compared LUR models for the Netherlands and Great Britain. They conclude that cautiousness is warranted when transferring LUR models to other regions or when transboundary models are built. On the other hand, the performance of models based upon common data or applied in very similar areas is only slightly worse than models optimized with local data. An additional, but limited, measurement campaign may help to improve transferability of a LUR model (Briggs et al., 2000; Poplawski et al., 2009). Dijkema et al. (2011) did a comparable exercise, but they put their focus on the correspondence between an urban and a regional LUR model in the Amsterdam region.

The aim of this study is to develop a LUR model for black carbon (BC) for the urbanized region of Flanders in Belgium. From the measurement campaign described in chapter 3, some fixed site measurements (21) are available, but these measurements were initially not intended to build a LUR model. More measurements (42) are available for a small part of the study area, namely for the city of Antwerp. These were collected in the framework of the HEAPS study with the explicit intention of building an urban LUR model (Van Poppel et al., 2012a). Using both available datasets, it will be explored whether an urban model is capable of predicting regional concentrations, or whether a regional model can predict concentrations in the urban area with sufficient accuracy. Combining both datasets, either in a new LUR model with 63 measurements, or in an urban or rural LUR model with recalibration of the coefficients, are options that will be studied.
BC is measured as this pollutant is highly relevant in the study area because of the high share of diesel cars. Moreover this pollutant is suspected to be responsible for important health effects (Janssen et al., 2011; WHO, 2012). Karner et al. (2010) showed that there are steep spatial gradients of BC near roads, so a model should be able to take these local differences into account.

4.1.2 MATERIALS AND METHODS

4.1.2.1 Study area

A regional LUR model will be developed for the Flemish Region, the northern part of Belgium, including the city of Brussels. The study area has a population of over 7 million, and a total area of 13,684 km². There are several larger cities in the area with 100,000 to 500,000 inhabitants and a population density of approximately 1000 inh/km², but also in between these cities the region is rather urbanized. The area is predominantly flat, and is situated along the North Sea in the west. Due to its central orientation in Europe, Flanders is one of the most important traffic hot spots in Western Europe. A peculiarity of the study area is the high share of diesel fueled passenger cars (62% in Belgium (NIS, 2010)). A separate LUR model is developed for the city of Antwerp (1 million inhabitants, approximately 1000 km²) one of the largest cities in the study area.

4.1.2.2 Air quality measurements

Two independent BC monitoring campaigns were conducted: (i) 42 sites in Antwerp, majority of sites in the city center and 2 sites on a suburban/rural location, measurements made in May-June 2011 and November-December 2011, during one week, all measurements in both seasons; (ii) 21 sites spread over Flanders, both in urban, suburban and rural environments, measurements made in May-July 2010 and December-February 2010-2011, during one week, repeated in a contrasting season on two locations. The first campaign was performed for the purpose of LUR modeling; the other covered residential locations of people participating in the personal monitoring study (chapter 3).

Sampling sites were selected based on traffic intensity and population density in the neighborhood. This was done to ensure enough spatial variation in measured concentrations (Lebret et al., 2000), while ensuring that measurement sites are representative for the area where the final model will be applied (Wang et al., 2012). Sites were grouped in four classes (TABLE 10; FIGURE 26): street locations (N=13), urban traffic locations (N=25), urban background locations (N=11) and rural locations (N=14). All BC monitors were located on the facade of mainly residential buildings or on street lights. Aethalometers, type AE51 (AethLabs, 2011), were placed in a weatherproof housing, and were mounted at a height of 2-3m. Aethalometers measured BC with a temporal resolution of 5-min. Measurements with highly loaded filters (attenuation>75) or with an error signal were omitted (similar to the approach reported in chapter 3.2).

TABLE 10: Definition of different location classes for sampling site selection

S	Street location	Adjacent road > 10000 vehicles/day
UT	Urban location with important traffic influence within 300m	Adjacent road < 10000 vehicles/day; less than 300m from a road > 10000 vehicles/day
UB	Urban location without important traffic influence within 300m	Adjacent road < 10000 vehicles/day; more than 300m from a road > 10000 vehicles/day; population density > 2000 inhabitants/km²
R	Rural location	Adjacent road < 10000 vehicles/day; more than 300m from a road > 10000 vehicles/day; population density < 2000 inhabitants/km²

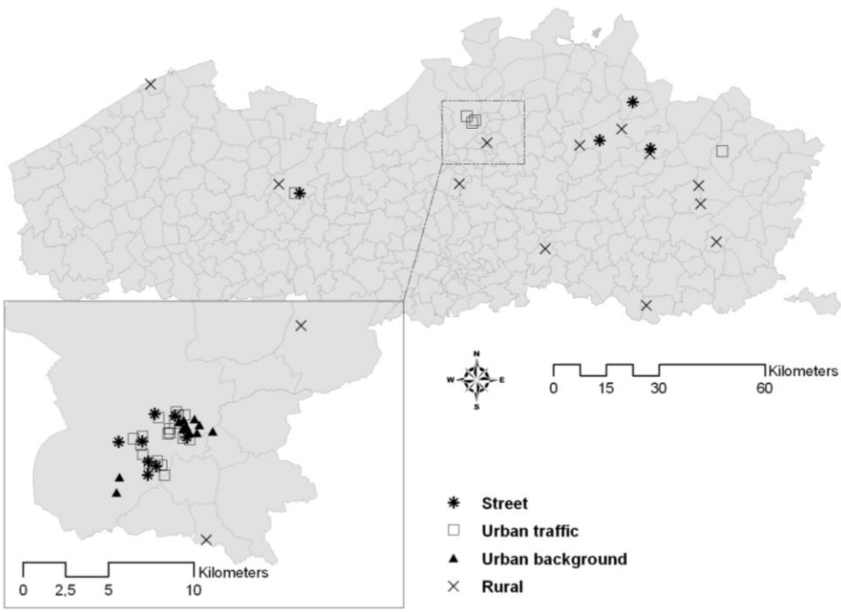


FIGURE 26: Study area with measurement campaign (i) (in extent rectangle) and campaign (ii) (large map), grouped by location class

Measurements were done simultaneously at 3 to 10 sites (mostly from a different type) during each week. Due to the limited availability of aethalometers, non-simultaneous measurements were re-estimated to account for relative differences in background concentrations. Continuous measurements at a fixed monitoring station were used for this, after checking for correspondence between the AE51 aethalometers and the MAAP (Multi-Angle Absorption Photometers) used in the official network ($R^2_{winter2010-2011}$=0.90; $R^2_{spring\ 2011}$=0.78; $R^2_{autumn\ 2011}$=0.89). TABLE 11 shows the BC concentrations measured on sites of the official air quality network throughout the monitoring periods. The correlation (R^2) between concentrations measured in 2010 and 2011 at 8 sites of the official air quality network was 0.91 with a slope of 1.03, indicating that year of sampling was probably not important (see also chapter 4.2). Seasonal measurements were rescaled to represent annual average concentrations, and the annual concentrations were used as input in the LUR models.

TABLE 11: Period averaged hourly BC concentrations [µg/m³] measured during our campaigns at all measurement stations in the official monitoring network in Flanders measuring BC (at least 90% available data based on hourly measurements, unless indicated differently)

Location ID	Description	Location type			Campaign (i)		Campaign (ii)	
			2010 (P25-P75)	2011 (P25-P75)	May 16, 2011 – June 28, 2011 (P25-P75)	November 15, 2011 – December 20, 2011 (P25-P75)	May 2, 2010 – July 8, 2010 (P25-P75)	December 9, 2010 – February 25, 2011 (P25-P75)
42M802	Antwerpen-Luchtbal	Industrial	2.4 (1.1-3.0)	2.6 (1.1-3.3)	1.65 (0.80-1.97)	3.52 (1.18-4.37)	1.89 (0.89-2.31)	2.95 (1.35-3.87)
42R801	Borgerhout	Urban	3.0 (1.4-3.9)	2.9 (1.3-3.7)	1.84 (0.96-2.33)	3.89 (1.41-5.22)	1.96 (1.04-2.44)	3.43 (1.65-4.59)
42R815	Zwijndrecht	Industrial	2.2 (0.9-2.8)	2.2 (0.9-2.8)	1.26 (0.66-1.54)	3.25 (0.92-3.72)	1.53 (0.76-1.99)	2.71 (1.13-3.62)
44M705	Roeselare-haven	Industrial	2.0 (0.8-2.5)	2.1 (0.7-2.8)	1.06 (0.40-1.38)	3.05 (0.88-3.64)	1.72 (0.82-2.20)	2.32 (0.95-3.05)
40AB01	Antwerpen-Boudewijnsluis	Industrial	2.0 (0.8-2.5)	2.2 (0.8-2.8)	1.28 (0.63-1.52)	2.88 (0.91-3.54)	1.38 (0.69-1.71)	2.60 (1.15-3.48)
40AL01	Antwerpen-Linkeroever	Suburban	1.8 (0.8-2.3)	1.9 (0.7-2.4)	1.03 (0.51-1.30)	2.64 (0.67-3.02)	1.30 (0.69-1.66)	2.19 (0.94-2.95)
40SZ01	Zaventem	Suburban	2.0 (0.9-2.6)	2.0 (0.9-2.5)	1.14 (0.64-1.44)	3.14 (1.08-3.51)	1.44 (0.73-1.77)	2.39 (1.15-3.25)
40R833	Stabroek	Suburban	1.7 (0.8-2.1)[a]	1.8 (0.8-2.3)	1.10 (0.61-1.39)	2.67 (0.83-3.23)	1.16 (0.53-1.47)[a]	2.14 (0.94-2.85)

[a] Measurements starting from June 8, 2010

4.1.2.3 Geographical covariates

Potential variables that may be correlated with measured BC concentrations include traffic variables, population variables, and land use variables (TABLE 12).

Traffic data is available from numerous datasets: detailed road maps, incomplete road maps with linked traffic intensities, road maps with information on heavy traffic. Detailed road maps are used to calculate total road length in buffers with different sizes. This map is also used to determine the distance to the nearest highway. Traffic intensity data originate from the activity-based traffic model FEATHERS built for Flanders and Brussels (Bellemans et al., 2010). Trips are considered as necessary if sequential activities are performed on different places. On a population level, these patterns result in origin/destination matrices that are assigned to a somewhat less detailed road network. Certain traffic variables are calculated for major roads only: these roads are defined as having a traffic intensity >10,000 veh/day. The AB model does not predict heavy traffic, but it includes heavy traffic from an external source as initial load on the road network, necessary for a realistic equilibrium assignment. Heavy traffic streams and fraction of heavy traffic on a route (based on automated traffic counts in the study area) are included in the LUR model development; these data are from a different source as the heavy traffic streams used in FEATHERS.

Population variables from two sources were used. The first dataset contains static address information, and it is based on information from the national bureau of statistics. Secondly, the dynamic population density is calculated. Dynamic population density is also an output of the AB model: personhours in every subzone are estimated for every hour of the day. This metric takes into account realistic population densities, e.g. in industrialized areas people are present during the day although no one lives there, at night population density is thus zero.

Land use variables are compiled from the CORINE land cover dataset (EEA, 2004), freely available from the EEA-website, and include build-up area, green area, industrial sites and (air)ports.

If applicable, variables were computed in several circular zones around measurement sites. The size of buffers for land use variables can be larger than buffers for traffic variables, as traffic factors have a local effect on air quality (Hoek et al., 2008; Su et al., 2009) and land use variables are often more representative of background concentrations. The size of the buffers should also be based upon the decay of the modeled pollutant, i.e. wider for PM_{10} and narrower for estimates of diesel (Ryan and LeMasters, 2007; Su et al., 2009). Our study area has a short coastline, and therefore a variable denoting the distance to the sea corresponding to higher wind speeds could be added (Henderson et al., 2007; Wilton et al., 2010). Because there is only one measurement site near the coastline, this was considered to be insufficient to catch this phenomenon. As the study area is predominantly flat, distance to the shore does not reflect height differences (Wilton et al., 2010). Distance to shore was therefore not included in the models.

TABLE 12: List of covariates

Variable name	Description	Buffer sizes
ROADLENGTH_XX	Total road length in a buffer with size XXm [m]	50, 100, 300, 500, 1000
D_HIGH	Distance to the nearest highway [m]	
D_HIGH_LT1000	Distance to the nearest highway < 1000m [a]	
ADDRESS_XX	Number of addresses in a buffer with size XXm	50, 100, 300, 500, 1000, 3000
HDRES_XX	High density residential in a buffer with size XXm (CORINE Class 111) [m²]	100[a], 300[a], 500[a], 1000, 3000, 5000
LDRES_XX	Low density residential in a buffer with size XXm (CORINE Class 112) [m²]	100, 300, 500, 1000, 3000, 5000
IND_XX	Industrial or commercial units in a buffer with size XXm (CORINE Class 121) [m²]	1000[a], 3000, 5000
Port_XX	Port area in a buffer with size XXm (CORINE Class 123) [m²]	3000, 5000
Airp_XX	Airport in a buffer with size XXm (CORINE Class 124) [m²]	1000[a], 3000, 5000
UrbGr_XX	Urban green in a buffer with size XXm (CORINE Classes 141, 142) [m²]	300[a], 500[a], 1000[a], 3000, 5000
NATURE_XX	Natural land in a buffer with size XXm (CORINE Classes 141, 142, 211, 231, 242, 243, 311, 312, 313, 322, 421, 511, 512, 522) [m²]	300[a], 500, 1000, 3000, 5000

PEOPLE_XX_WKDYx	Number of people in a buffer with size XXm at hour x (WKDY weekday; WKND weekend)	50, 100, 300, 500, 1000, 3000
Q_NEAR_WKDYx	Traffic intensity (private cars) on the nearest road at hour x (WKDY weekday; WKND weekend) [veh/h]	
DIST_NEAR	Distance to the nearest road [m] [b]	
D_NEAR1	1 / Distance to the nearest road [1/m] [b]	
D_NEAR2	1 / (Distance to the nearest road)² [1/m²] [b]	
QD_NEAR1_WKDYx	Traffic intensity (private cars) on the nearest road at hour x / distance to the nearest road [b]	
QD_NEAR2_WKDYx	Traffic intensity (private cars) on the nearest road at hour x / (distance to the nearest road)² [b]	
TRAFLOADXX_WKDYx	Sum of (traffic intensity (private cars) at hour x * road length) in a buffer with size XXm	50, 100, 300, 500, 1000
Q_NEAR_MAJOR_WKDYx	Traffic intensity (private cars) on the nearest major road at hour x (WKDY weekday; WKND weekend) [veh/h] [c]	
DIST_NEAR_MAJOR	Distance to the nearest major road [m] [c]	
D_NEAR_MAJOR1	1 / Distance to the nearest major road [1/m] [c]	
D_NEAR_MAJOR2	1 / (Distance to the nearest major road)² [1/m²] [c]	
TRAFLOADMAJOR_XX_WKDYx	Sum of (traffic intensity on major roads (private cars) at hour x * road length of major roads) in a buffer with size XXm [c]	50[a], 100[a], 300, 500, 1000
Q_NEAR_HEAVY	Traffic intensity (heavy traffic) on the nearest road [veh/h] [d]	
QD_NEAR1_HEAVY	Traffic intensity (heavy traffic) on the nearest road / distance to the nearest road [d]	
QD_NEAR2_HEAVY	Traffic intensity (heavy traffic) on the nearest road / distance to the nearest road [d]	
TRAFLOAD_XX_HEAVY	Sum of (traffic intensity (heavy traffic) * road length) in a buffer with size XXm [d]	50[a], 100, 300, 500, 1000
TRAFLOADHV_FRACTION_XX	Fraction of heavy traffic in a buffer with size XXm [e]	100, 300, 500, 1000
TRAFLOADHV_FRACTION_XX_2	Fraction of heavy traffic squared in a buffer with size XXm [e]	100, 300, 500, 1000

[a] Variable is indicator variable
[b] Minor roads are not included in the traffic intensity network. If the distance between the nearest road of the traffic intensity network and the detailed network is larger than 20m, the distance to the detailed network is selected as the distance to the nearest road. Traffic intensity on a road with unknown intensities is set to 500 veh/day. Hourly traffic intensities are calculated based on 500 veh/day and hourly factors as defined in http://www.tmleuven.be/project/car/Handleiding_CAR-Vlaanderen_v2.0.pdf (In Dutch).
[c] Major roads are roads with annual average daily traffic of private cars > 10,000 vehicles
[d] Heavy traffic only in Flanders (not in Brussels)
[e] All data originate from the multimodal traffic model (traffic load of all traffic and traffic load of heavy traffic)

4.1.2.4 LUR model development

Initially, the correlation between all potential predictor variables was calculated; if two variables were highly correlated ($|r|>0.95$), one of the variables was omitted to prevent multicollinearity in regression models (Henderson et al., 2007). Variables correlated ($|r|>=0.6$) with the most significant variable in one category, were omitted, again to prevent multicollinearity issues in further stages of the LUR model development. Remaining variables were entered in a supervised forward stepwise regression; variables were selected based on the adjusted R^2. Only variables that are likely to contribute to BC concentrations were allowed in the model. It would be possible to develop statistically significant models but that clearly violate physical laws. This was avoided by choosing realistic covariates and by forcing selected coefficients to be consistent with a priori assumptions on the direction of effect (Johnson et al., 2010). Additional criteria that needed to be met include: the variable had to contribute at least 1% to the adjusted R^2; and the direction of effect of variables already selected should not change when including an additional variable (Dijkema et al., 2011; Eeftens et al., 2012). One variable could be included in the final model in multiple buffer sizes; it was preferred to include the original nested buffers rather than using concentric adjacent rings (von Klot, 2011). Finally, all variables included in the model needed to have significant t-statistics ($\alpha=0.05$) or they were removed from the model. Variance inflation factors (VIFs) were calculated to check for multicollinearity between variables (Beelen et al., 2013; Eeftens et al., 2012; Johnson et al., 2010). This procedure is similar to methods used in previous LUR studies (Eeftens et al., 2012; Henderson et al., 2007).

In this chapter we discuss seven different LUR models: M1 to M7. Independent models were developed for the 2 datasets: a model based on repeated measurements from the first campaign on 42 sites in and around the city of Antwerp (M1); and a model based on 21 measurements from the second campaign spread over Flanders (M4). Additionally, both of these models were recalibrated with the results of the other measurement campaign by keeping the significant variables, but by reassessing the coefficients (M2, M5), in analogy with methods used by Briggs et al. (2000) and Poplawski et al. (2009). Instead of using only the urban or regional dataset for recalibration of the model, both models were also reassessed using all 63 measurements as all those points are geographically situated in same the study area (M3, M6). The final model (M7) considered measurements from both campaigns as interchangeable and this model was built using all 63 measurements. All models developed (M1-M7) are validated against three datasets: 42 measurements from campaign (i), 21 measurements from campaign (ii), and 63 measurements from both campaigns together. Leave-one-out cross-validation (LOOCV) is used to validate single models using (n-1) measurements by comparing the measurement at the left-out site with the model prediction; this is repeated n times (Brauer et al., 2003; Hoek et al., 2008).

4.1.3 RESULTS

In the first campaign, including mostly urban sites, an annual average concentration of 2452 (±528) ng/m³ was measured. In the second campaign, consisting of measurements at 21 urban and regional sites in Flanders, the average concentration was lower: 1735 (±807) ng/m³ after rescaling. Minimum and maximum values for selected predictor variables are mostly comparable between the two campaigns (TABLE 13). As the second campaign also included several rural sites, the lowest concentrations are measured in this second campaign; the maximum concentration is comparable in both campaigns.

TABLE 13: Minimum and maximum values for a selection of dependent and independent variables in both measurement campaigns (TRAFLOAD_50_HEAVY and UrbGr_1000 are indicator variables).

Variable	Campaign (i) mostly urban sites (N=42)	Campaign (ii) regional and urban sites (N=21)
Measured BC concentrations	[1522; 4184]	[846; 3865]
TRAFLOAD_50_HEAVY	[0; 1]	[0; 1]
TRAFLOADHV_FRACTION_100	[0; 0.28]	[0; 0.08]
ROADLENGTH_1000	[20,405; 64,003]	[9809; 62,952]
ADDRESS_50	[1; 53]	[1; 47]
UrbGr_1000	[0; 1]	[0; 1]
DIST_NEAR	[1; 100]	[1; 36]

The first model M1 is a classical LUR model using repeated measurements from 42 sites, rescaled to represent annual average concentrations (campaign (i)). The R^2 of the model is 0.73, with a LOOCV R^2 of 0.46 (TABLE 14). This model consists of 4 significant variables, of which 3 are traffic variables (road length within 1000m, and heavy traffic variables in smaller buffers). There is no spatial autocorrelation in the residuals (Moran's I=-0.12, p=0.21). Models M2 and M3 retain the significant variables of M1, but coefficients are re-estimated. This results in a negative effect of 'fraction of heavy traffic' which is counter-intuitive because increased heavy traffic emissions should result in increased concentrations. The heavy traffic-variables in model M2 lose their significance at the 95% level. Heavy traffic in 50m and the 'fraction of heavy traffic in 100m' both have inflated variance inflation factors (VIF=4.17) in model M2. Residuals show a rather dispersed pattern (Moran's I=-0.30, p=0.20). In model M3 all variables have the expected direction of effect. The R^2 and adjusted R^2 are similar to model M1, but the RMSE is larger due to the larger spread in concentrations. LOOCV R^2 is 0.63, which is higher than the validation R^2 of model M1.

Model M4 is built using measurements at 21 sites from campaign (ii). Compared to M1, this model includes a land use variable, urban green in 1000m, and two traffic variables (road length within 1000m and fraction of heavy traffic in 100m; see TABLE 14). VIFs of the variables in models M4, M5 and M6 are well below 2. Model M4 shows a borderline significant negative spatial autocorrelation of the residuals, suggesting a dispersed pattern of the residuals. M4 has high R^2, adjusted R^2 and LOOCV R^2 values. The R^2 of model M5 is much lower (R^2=0.54). All variables in models M5 and M6 have the expected direction of effect, although the effect sizes differ between models.

Model M7, using 63 measurements from both campaigns, again consists of 4 variables; most of them were also present in previous models. This model only includes traffic variables, although total length of roads in 1km is expected to be more representative for urbanization than for traffic. All variables have VIFs below 1.4 and the residuals show no spatial autocorrelation (Moran's I:-0.06, p=0.53). The difference between the R^2 and the validation R^2 is small; this also holds for the RMSE.

To further evaluate the performance of the LUR models, measured and modeled concentrations are compared. Models M1 to M7 are applied to all 63 points, and to the 42 measurements of the first campaign and the 21 measurements of the second campaign separately (TABLE 15, FIGURE 27). Models estimated with 21 measurements are less efficient in estimating concentrations for the other 42 sites, than for the 21 sites used to select significant variables. Model M1, based on 42 mostly urban sites, cannot predict concentrations for urban and regional sites in Flanders with enough reliability (R^2=0.50). Model M2, with urban variables and recalibrated using measurements of the regional measurement campaign, has a low R^2 of 0.24 when reapplied to the 42 urban sites of the first campaign. It seems that the three significant variables from the regional model using 21 urban and rural sites, cannot capture enough variability necessary to accurately predict concentrations at many of the urban sites. Model M7, estimated using measurements on 63 sites, performs well in total and in both subsets.

TABLE 14: Overview of 7 LUR models built to model BC concentrations in Flanders

		R^2	Adj R^2	RMSE	LOOCV R^2	LOOCV RMSE	LUR model [a]
M1	Urban LUR model (42 sites)	0.73	0.70	290.15	0.46	384.47	c_{BC} = 1242 + (619*TRAFLOAD_50_HEAVY) + (3960*TRAFLOADHV_FRACTION_100) + (0.015*ROADLENGTH_1000) + (8.8*ADDRESS_50)
M2	Urban LUR model recalibrated with 21 regional sites	0.82	0.78	380.02	0.63	486.37	c_{BC} = 156 + (709*TRAFLOAD_50_HEAVY) - (2396*TRAFLOADHV_FRACTION_100) + (0.066*ROADLENGTH_1000) - (31.7*ADDRESS_50)
M3	Urban LUR model recalibrated with 63 sites	0.74	0.72	375.59	0.63	429.82	c_{BC} = 641 + (437*TRAFLOAD_50_HEAVY) + (4753*TRAFLOADHV_FRACTION_100) + (0.031*ROADLENGTH_1000) + (2.47*ADDRESS_50)
M4	Regional LUR model (21 sites)	0.88	0.85	309.24	0.76	387.17	c_{BC} = 385 + (0.052*ROADLENGTH_1000) - (740*UrbGr_1000) + (14214*TRAFLOADHV_FRACTION_100)
M5	Regional LUR model recalibrated with 42 urban sites	0.54	0.50	372.67	0.44	390.73	c_{BC} = 1286 + (0.022*ROADLENGTH_1000) - (144*UrbGr_1000) + (7023*TRAFLOADHV_FRACTION_100)
M6	Regional LUR model recalibrated with 63 sites	0.71	0.69	397.12	0.67	407.35	c_{BC} = 757 + (0.034*ROADLENGTH_1000) - (212*UrbGr_1000) + (7865*TRAFLOADHV_FRACTION_100)
M7	Urban and regional site LUR model (63 sites)	0.77	0.75	356.84	0.72	373.88	c_{BC} = 812 + (0.031*ROADLENGTH_1000) + (4569*TRAFLOADHV_FRACTION_100) + (394*TRAFLOAD_50_HEAVY) - (8.06*DIST_NEAR)

[a] Traffic load of heavy traffic in a buffer with radius 50m – indicator variable (TRAFLOAD_50_HEAVY), Fraction of traffic load that is heavy traffic in a buffer with radius 100m (TRAFLOADHV_FRACTION_100), Total road length in a buffer with radius 1km (ROADLENGTH_1000), Number of addresses in a buffer with size 50m (ADDRESS_50), Urban green in a buffer with size 1000m (UrbGr_1000), Distance to the nearest road (DIST_NEAR)

TABLE 15: Comparison of 7 different LUR models applied to regional and/or urban locations (Y=modeled; X=measured)

		Campaign (i) mostly urban sites (N=42)		Campaign (ii) regional and urban sites (N=21)		Campaign (i) + (ii) regional and urban sites (N=63)	
		Regression function	R^2	Regression function	R^2	Regression function	R^2
M1	Urban LUR model (42 sites)	Y = 0.73x + 667	0.73	Y = 0.39x + 1320	0.50	Y = 0.57x + 1042	0.67
M2	Urban LUR model recalibrated with 21 regional sites	Y = 0.77x + 919	0.24	Y = 0.82x + 308	0.82	Y = 0.96x + 329	0.52
M3	Urban LUR model recalibrated with 63 sites	Y = 0.77x + 570	0.67	Y = 0.57x + 737	0.70	Y = 0.74x + 572	0.74
M4	Regional LUR model (21 sites)	Y = 1.17x - 84	0.50	Y = 0.88x + 217	0.88	Y = 1.12x - 35	0.68
M5	Regional LUR model recalibrated with 42 urban sites	Y = 0.54x + 1131	0.54	Y = 0.40x + 1267	0.81	Y = 0.52x + 1147	0.69
M6	Regional LUR model recalibrated with 63 sites	Y = 0.68x + 791	0.52	Y = 0.57x + 746	0.82	Y = 0.71x + 650	0.71
M7	Urban and regional site LUR model (63 sites)	Y = 0.83x + 420	0.69	Y = 0.60x + 705	0.77	Y = 0.77x + 516	0.77

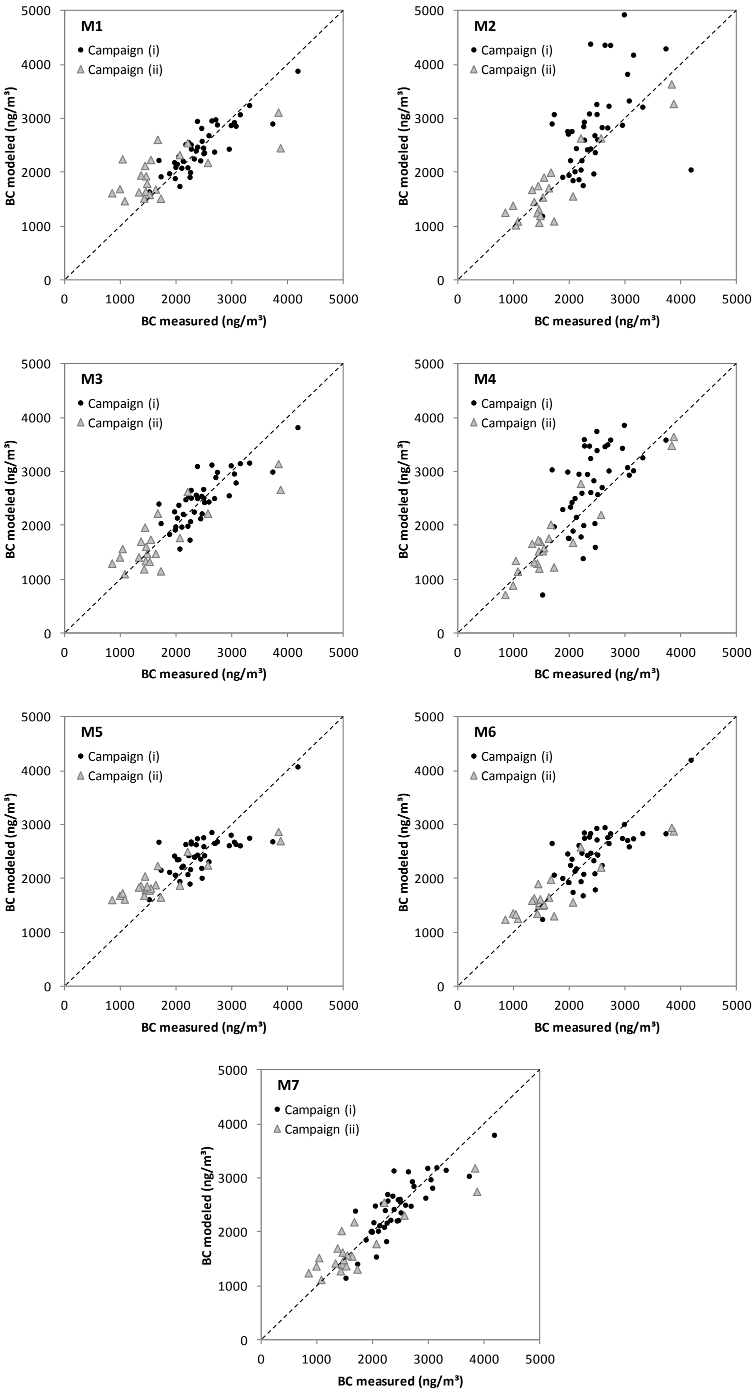


FIGURE 27: Results of 7 different LUR models applied to campaign (i) (42 mostly urban sites) and campaign (ii) (21 regional and urban sites)

4.1.4 DISCUSSION

In this chapter, 7 different land use regression models for BC were developed and validated. All models, M1-M7, were estimated using either 42 mostly urban measurements (campaign (i)), 21 regional and urban measurements (campaign (ii)), or both datasets together. It was tested whether the LUR model with mostly urban measurement sites was able to predict concentrations in other cities and on more rural locations. Alternatively, it was examined whether measurements at just 21 sites in a relatively large area, would suffice to predict concentrations in a small urban zone located inside the larger study area.

The transferability of LUR models from one study area to another is enhanced when covariates originate from the same dataset in the different areas (Hoek et al., 2008; Poplawski et al., 2009; Vienneau et al., 2010). In our study area, all variables met this precondition (traffic intensities, road network, land use, address density); moreover the urban area is geographically situated inside the regional study area. BC was measured using the same study protocol in both measurement campaigns: outdoor measurements with aethalometers type AE51 at the facade of residences, for 7 consecutive days, on a 5-min time resolution. A drawback of the second sampling campaign is the limited number of locations (N=21) and the fact that only on 2 locations the weekly measurement was repeated in a different season. In the first campaign with many urban sites, all locations were measured twice: once in spring/summer and once in autumn/winter. The first campaign was executed between May 2011 and December 2011, the second campaign between May 2010 and February 2011; therefore attention should be paid to potential changes in background concentrations over the years, although this is not considered a major issue in this study (TABLE 11). Taking these factors into account, it should be possible to merge both datasets into one larger dataset with 63 observations.

A list of potential covariates was composed with traffic, land use and population variables. Variables were selected based on a literature review on significant variables in LUR models for BC, black smoke, elemental carbon, or absorbance of PM (TABLE 16). Traffic variables are dominant in most published LUR models;

a population variable is often included; whereas land use variables only sporadically turn out to be significant predictors. Meteorological variables are included in one model (Gryparis et al., 2007), 'season' in another one (Levy et al., 2010). Traffic variables are often included at short distances of 50-100m, as the effects of traffic are very local, especially in Europe (Hoek et al., 2008). Traffic variables in larger buffers will be more representative of the degree of urbanization rather than of local traffic (Beelen et al., 2013). Performance of the models reported in literature ranges from R^2=0.39 to R^2=0.82; the results achieved in this study are at the higher end of this interval. Most models are based on 40 or less measurement sites, while several models are built with measurements on just 25 sites. A minimum of 40-80 sites has been suggested for estimating robust LUR models depending on the size and diversity of the study area (Hoek et al., 2008); with a lower number of measurement sites the LOOCV R^2 will be an overestimation of the hold-out validation R^2 (Basagaña et al., 2012; Wang et al., 2012). To minimize problems of over fitting, the set of potential covariates should be limited, or an a priori selection should be made, e.g. by removing highly correlated variables (Basagaña et al., 2012; Eeftens et al., 2012). Preference should be given to models with at least 40 measurement sites, in this study this excludes models M2, M4, M5, and M6.

Model M1 was capable of predicting concentrations at sites outside of the urban region with an acceptable accuracy (R^2=0.50, compared to LOOCV R^2=0.46). Unfortunately, lower concentrations, in this case on rural locations, are overestimated because of the absence of rural sites in campaign (i). This was also reported by Dijkema et al. (2011) when applying a city specific LUR model for Amsterdam to rural sites. The use of model M1 would lead to an overestimation of BC concentrations in rural areas, affecting lots of people in Flanders. Utilizing campaign (ii) to validate model M1 or campaign (i) to validate model M4 can be considered as an external and independent validation, but while taking into account that the validation dataset consists of sites with partly different characteristics than the calibration dataset. Local recalibration of models M1 and M4 with respectively measurements from campaign (ii) and campaign (i) (M2 and M5) improves the performance of the original model in the new study area. In this study though, a model that performs well in both urban and regional areas, and can reproduce measurements from both measurement campaigns is necessary. Multiple variables in models M2, M3 and M5, lose their significance at the 95% level; this corresponds to observations from Poplawski et al. (2009) when transferring a LUR model from Vancouver to Seattle and Victoria. Model M3 and model M7 are very much alike, with M7 having slightly better performance and validation criteria. In internal LOOCV, the percentage of explained variability was very close to the model R^2, suggesting good applicability of the model to unmeasured locations. The variable 'Addresses within 50m' is not significant anymore in model M3, and is replaced by 'distance to the nearest road' in model M7 resulting in a better model.

In our study set up, both campaigns were geographically overlapping, but one campaign was centered around an urban area, while the other also contained rural and regional sites. Ideally, one integrated monitoring campaign in Flanders with at least 40 measurements at representative sites should be aimed at. However purpose-designed monitoring campaigns are expensive and using available data proved to be a good alternative. Measurements took place in consecutive years, but background concentrations remained stable over two years. After rescaling of the non-simultaneous measurements, there was no apparent reason why not to combine measurements from both measurement campaigns into one dataset. Developing a new LUR model based on 63 measurements is therefore preferred over the use of an existing urban LUR model. It was already shown in literature that transferability is unpredictable and other variables might be more predictive for a new study area.

In the remainder of this book we will continue the work towards an integrated exposure model by designing a LUR model applicable to the whole study area of Flanders.

TABLE 16: Land use regression models for BC or correlated pollutants ($PM_{2.5}$ absorbance – $PM_{2.5}$ abs, Elemental Carbon - EC, Black Smoke - BS)

Author	Pollutant	Number of measurement sites	Study area	Predictor variables	Model performance
(Gryparis et al., 2007)	BC	30 (BC outdoor) + 15 (BC ambient) + 23 (EC outdoor)	Boston, Massachusetts, USA	- Meteorology (e.g., wind speed, temperature) - Traffic density within a 100-m radius - Population density - Distance to nearest major roadway - Percent urbanization - BC concentrations at a central monitor - Other (e.g., year, day of the week, longitude, latitude)	R^2=0.82
(Brauer et al., 2008)	BC	39	Vancouver, British Columbia, Canada	- Length of major roads within a 100-m radius - Distance to nearest highway - Area of industrial land within a 750-m radius	R^2 = 0.56
(Brauer et al., 2003)	$PM_{2.5}$ abs	40	Netherlands	- Number of high-traffic roads in 250-m buffer - Household (address) density in 300-m buffer - Distance major road - Region	R^2=0.81
		40	Munich, Germany	- Traffic load (50-250m) - Traffic load (50m) - Population density (300m) - Population density (300-5000m)	R^2=0.67
		42	Stockholm County, Sweden	- Traffic flow on nearest road - Population density (1000-5000m)	R^2=0.66
(Hochadel et al., 2006)	$PM_{2.5}$ abs	40	North Rhine-Westphalia, Germany	Model (a): homogenous rural area - Daily traffic in 250-m radius - Maximum traffic intensity in 50-m radius - Distance to highway	R^2=0.65
				Model (b): range of urbanization degrees & traffic densities - Daily traffic heavy vehicles (100m-10km) - Daily traffic in 100m radius - Distance to highway	R^2=0.82

(Morgenstern et al., 2007)	$PM_{2.5}$ abs	40	Munich, Germany	- Household density (2500-5000m) - Distance to nearest federal roads - Land coverage factor (100-250m) - Length of county roads (0-1000m)	R^2=0.42
(Henderson et al., 2007)	$PM_{2.5}$ abs	25	Vancouver, British Columbia, Canada	- Length of highways in 1000-m radius - Length of major roads in 100-m radius - Distance to highway - Open area in 500-m radius - Truck density in 1000-m radius	R^2=0.39 (model with first 4 variables) R^2=0.41 (model with 'truck density' variable)
(Levy et al., 2010)	$PM_{2.5}$ abs	44	Boston, Massachusetts, US	- ln(EC) concentrations at a central monitor * cooling season - Roadway length in 200-m radius * % hours of still winds - Cooling season	R^2=0.52
(Eeftens et al., 2012)	$PM_{2.5}$ abs	20 (for 18 study areas) 40 (for 2 study areas)	Europe	- Traffic load - Traffic load of heavy traffic - Road length - Land use	R^2=0.56-0.97
(Clougherty et al., 2013)	$PM_{2.5}$ abs	155	New York City, New York, US	- Concentration at a reference site - Truck traffic in 1-km radius - Number of oil-burning units in 200-m radius - Area of industrial land within a 1-km radius - Kernel-weighted traffic within 100m	R^2=0.65
(Beelen et al., 2007)	BS	23	Netherlands	- (Number of inhabitants/1000) in a 1000-m radius - Region - Traffic intensity in a 100-m radius	R^2=0.59
(Carr et al., 2002)	EC	34	Munich, Germany	- Traffic intensity in 50-m radius - Traffic intensity between 50m and 300m - Traffic jam in 50-m radius - Traffic jam between 50m and 300m	R^2=0.80
(Ryan et al., 2007)	EC	24	Cincinnati, Ohio, USA	- Elevation - Avg daily truck count on major roads within 400-m radius - Length of bus routes within 100-m radius	R^2=0.75

4.2 MODELING TEMPORAL AND SPATIAL VARIABILITY OF TRAFFIC-RELATED AIR POLLUTION: HOURLY LAND USE REGRESSION MODELS FOR BLACK CARBON

This chapter is based on:

Dons, E., Van Poppel, M., Kochan, B., Wets, G., Int Panis, L., 2013. Modeling temporal and spatial variability of traffic-related air pollution: Hourly land use regression models for black carbon. Atmospheric Environment 74, 237-246.

4.2.1 INTRODUCTION

Black carbon (BC) is the dark carbonaceous component of particulate matter (PM). It was recently implicated in global warming (Jacobson, 2002; Shindell et al., 2012) and as a key component in the health effects associated with PM exposure (Janssen et al., 2011; von Klot et al., 2011). Exposure to BC is very variable in space and time and specific locations and activities may contribute a large share of total exposure (see chapter 3.1 and 3.2). Hence epidemiological studies of BC need to take into account exposure at different indoor and outdoor locations other than the home address. Using atmospheric dispersion models to achieve this, comes at a large cost in times of data collection, model set-up and computing times (Beckx et al., 2009c; Beckx et al., 2009d) and given the need for an extremely high spatial resolution, only few have been built for BC (Lefebvre et al., 2011).

Land use regression (LUR) modeling is a convenient statistical technique often used to determine exposure to air pollutants in epidemiological studies. In short, a LUR model tries to predict concentrations measured at 20 to 100 monitoring stations, by using traffic, land use or population density parameters as explanatory variables in a multiple linear regression model (Briggs et al., 1997; Hoek et al., 2008; Jerrett et al., 2005a). Once the model is developed, concentrations can be determined for any other location in the study area. Most LUR models consider annual average concentrations.

In epidemiological studies, time-activity diaries are used in combination with LUR models to estimate exposure: concentrations are determined at each location visited throughout the day, weighted for the time spent at each location (Ryan et al., 2008; Setton et al., 2011). A critique to this approach is that, as concentrations vary over a day, 24h-averages are used instead of real-time concentrations. Exposure at home tends to be overestimated (because people are at home during night hours) and exposure at work locations is underestimated (people are at work during the day) (Dhondt et al., 2012a). This is especially relevant for traffic-related air pollutants that are highly variable in time and space, such as BC or NO_2 (nitrogen dioxide). The aim of this study is to build hourly LUR models for BC by using both high-resolution monitoring and high-resolution covariates to better capture exposure at different times and locations. In the recent past, efforts have been made to incorporate a time-resolution into LUR models. The most straightforward way being the application of a temporal trend observed at a fixed monitoring station to the LUR predictions (Gan et al., 2011; Nethery et al., 2008b; Slama et al., 2007). Secondly, a temporal resolution can be introduced into a LUR model by including a dummy variable reflecting different time periods (e.g. season (Clougherty et al., 2009; Crouse et al., 2009), month (MacIntyre et al., 2011), weekday/weekend (Noth et al., 2011), day of the week (Maynard et al., 2007)). These approaches assume the same temporal trend at every site, whereas it is expected that during the day BC concentrations at street-locations will fluctuate much more than concentrations at background locations. Another approach is to recalibrate an existing LUR model with new measurements at the same monitoring stations to reflect different time periods (Mölter et al., 2010b; Wang et al., 2013), or to allow that model variables and coefficients change over different time periods and build several unique models (Gulliver et al., 2011b).

4.2.2 MATERIALS AND METHODS

4.2.2.1 BC measurement data

Continuous BC measurements were performed at 63 fixed locations on a 5-min time base. Measurement sites were chosen at building facades or on street lighting (height approximately 2-3m), using aethalometers model AE51 (AethLabs, 2011) in a weatherproof housing (appendix FIGURES A22 and A23). Twice or three times a week filters were replaced to prevent saturation of the filter strip. During sampling site selection, sites were grouped in 4 classes to ensure enough variation in expected concentrations (13 street sites, 25 urban traffic sites, 11 urban background sites, 14 rural sites; definitions in TABLE 10 (chapter 4.1). A first set of measurements consisted of BC measurements at 21 sites, spread over the Flemish region in Belgium (FIGURE 28). Both urban and rural sites were included in this monitoring campaign. Measurements were made either in May-July 2010, or in December 2010-February 2011. On two sampling locations measurements took place in both seasons. A second campaign consisted of measurements on 40 urban and 2 suburban locations (FIGURE 28). The measurements lasted for one week, and were repeated in a contrasting season (May-June 2011, November-December 2011). The second campaign was performed for the purpose of LUR modeling; the other covered residential locations of people participating in a personal monitoring study.

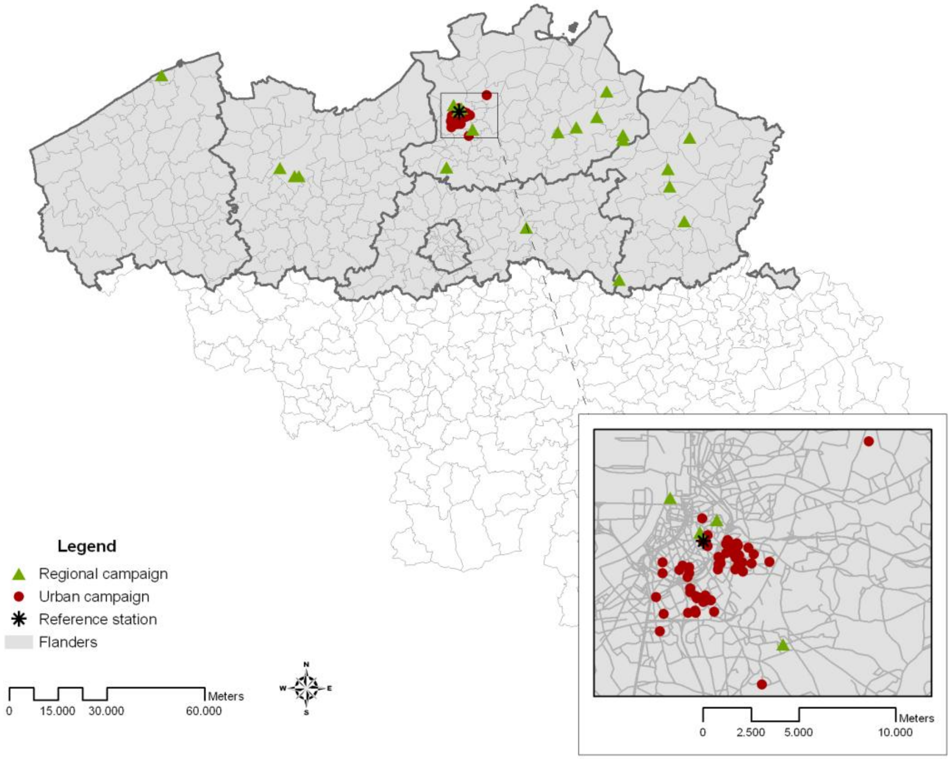


FIGURE 28: Study area Flanders with 63 measurement sites. Red dots indicate the 42 locations of the urban measurement campaign; the 21 locations of the regional measurement campaign are indicated by green triangles.

Measurements are too few to derive average concentrations over each hour of the week (i.e. 168 hours);local events would significantly influence the hourly concentration. Therefore 5-min measurements were aggregated to 24 hourly values for weekdays and 24 values for weekend days (appendix FIGURE A24). Because not all measurements were done simultaneously, hourly measurements from a reference monitor from the official air quality network were used to rescale the measurements based on variations in background concentrations (ratio of the measurement hour and the average concentration on that hour during the whole measurement period (measurement period either Summer and Winter 2010, or Summer and Winter 2011)). Air quality in 2010 and 2011, and in both summer and winter seasons was comparable; this was checked by consulting concentrations measured at 8 monitoring stations of the official air quality network (TABLE 11, chapter 4.1). Seasonal measurements were not analyzed separately, and only one annual average or hourly average concentration per location was used as dependent variable in the model development stage, summing to 63 sites.

4.2.2.2 GIS covariates

Both static and dynamic variables are used in this study to build hourly LUR models.

- Traffic variables: Hourly traffic streams are derived from the activity-based model for Flanders, FEATHERS (Bellemans et al., 2010). Hourly values are summed to derive daily traffic volumes. Heavy duty traffic is not included in FEATHERS; traffic streams of heavy goods vehicles originate from the Flemish multimodal traffic model (base year 2007). Detailed road maps are used to calculate total road length in buffers.
- Population variables: Population density and address density are two distinct layers. Address density is a point layer with static information on addresses. Population density originates from the FEATHERS model: for each subzone the size of the population present in that subzone is modeled for each hour. Population in buffers is then area-weighted.
- Land use variables: CORINE land cover data (EEA, 2004) are used for land use variables (base year 2000).

Altitude / elevation data are not collected since there are no major height differences in Flanders (<150m). Meteorological variables were not included because they are not readily available in the study area, although some studies indicate that meteorology can cause a small but significant improvement in LUR models (Arain et al., 2007; Wilton et al., 2010). Smaller buffer sizes are chosen for traffic variables and larger buffer sizes for land use variables. Variables are required to have values different from zero for at least 45% of locations; otherwise the variable is transformed into a binary indicator variable. If over 90% of the values for a variable equal zero, that variable is deleted. A complete list of predictor variables is included in TABLE 12 (chapter 4.1). All GIS calculations were performed in ArcGIS 9.3.

4.2.2.3 Model development algorithm

There is not yet a universal procedure to develop a LUR model, meaning that another procedure could lead to a more predictive model. The procedure followed in this study, is a combination of methods used by Henderson and others (2007) and Eeftens and others (2012). In step 1 the correlation between predictor variables is calculated; variables that are highly correlated are omitted ($|r|>0.95$), i.e. the variable that shows the least correlation with the measured concentration. Secondly, the independent variable with the highest correlation with the dependent variable in each sub-category is identified. If other variables in the same sub-category are correlated ($|r|\geq 0.6$) with the most highly ranked variable, they are removed. Remaining variables are entered in a supervised forward stepwise regression, with selection criterion 'adjusted R^2'. Other criteria that need to be met: the variable has to contribute at least 1% to the adjusted R^2; coefficients are consistent with a priori assumptions (direction of effect); the direction of effect of variables already selected does not change (Dijkema et al., 2011; Eeftens et al., 2012). As a final step, all variables with insignificant t-statistics ($\alpha=0.05$) are removed. The final equation is of the form $BC = \beta_0 + \beta_1X_1 + \beta_2X_2 + \ldots + \beta_xX_x$.

Hourly LUR models are developed using multiple approaches. A first option is to include dummy variables in a LUR model representing different hours of the day (DM – Dummy model). Therefore the input dataset is transformed into a dataset with 3005 cases (63 sites x 48h, excluding 19 missing hours). Weekday hour 0 is chosen to be the reference hour and not included in the model, the other 47 hourly dummy variables are forced into the model. The model is then estimated according to the selection criteria used for the annual model. The second method to derive hourly LUR models is to use static covariates and hourly concentration measurements, but building different hourly models instead of one annual model. Models can be developed by forcing the predictor variables from the annual model into the hourly models and by using the hourly concentration measurements for the prediction of the coefficients (HM1 – Hourly model 1). This technique was already proposed by Briggs and others (Briggs, 2005; Briggs et al., 2000) to develop LUR models for different seasons, and applied by Mölter and others (2010b). A similar approach develops models, independent of each other, by changing the dependent variable to reflect different hours of the day; but, in contrast with the previous method, significant variables may vary from hour to hour (HM2 – Hourly model 2). If dynamic variables (i.e. variables that change on an hourly basis) are available, they can be used instead of the static variables (HM3 – Hourly model 3). Predictor variables representing land use categories or road length will be constant over different hours; traffic intensity and population density will change during the day and can be predicted by an activity-based model (Beckx et al., 2009d). In this study we use predictions of the Flemish activity-based model FEATHERS (Bellemans et al., 2010). Instead of just replacing each static variable with its dynamic complement, the LUR model was estimated all over again to permit changes in other variables of the model or to omit the dynamic variable if it is no longer significant. Lagged effects, i.e. the impact of emissions of a previous hour on current concentrations, were also considered.

SAS 9.2 was used for model development.

4.2.3 RESULTS

4.2.3.1 Environmental sampling

Observed annual average concentrations ranged from 846 ng/m³ to 4184 ng/m³, with a mean value of 2213 ng/m³ (TABLE 17). The highest concentration was measured during the urban measurement campaign, and the lowest concentration during the regional campaign. The street site with the lowest mean concentration (1034 ng/m³) is further away (approximately 20m) from the adjacent road than other street sites, which explains lower concentrations. One urban traffic site with elevated concentrations is identified to have a high fraction of heavy traffic from a nearby factory passing through the street, yet the total traffic intensity is not exceeding 10,000 veh/day. There is significant spatial autocorrelation with elevated concentrations clustered in urban areas (Moran's I = 0.21; $p<0.05$).

TABLE 17: Results of BC sampling [ng/m³] at four different location types

	N	Mean	Median	StdDev	Min	Max
Street	13	2654	2713	766	1034	3828
Urban Traffic	25	2417	2363	660	1327	4184
Urban Background	11	2224	2212	297	1884	2954
Rural	14	1433	1464	308	846	2069

Valid hourly concentrations are obtained at all 63 locations on weekdays (appendix TABLE A10). On one site, measurements are missing for weekend days from midnight through 5 p.m., on another location measurements are missing for weekend days at 11 p.m.; both gaps are caused by instrument failure. Highest hourly concentrations are measured on street sites, with concentrations of nearly 10,000 ng/m³. The variability of spatial patterns during the day was explored and it appeared that the diurnal pattern changes when considering different locations (appendix FIGURE A25): a traffic site has high concentrations on traffic peak hours and low background concentrations at night (camelback pattern); a rural site has uniformly low concentrations. This also appeared from the Spearman rank correlation coefficients: on weekdays between 7 a.m. and 10 p.m. correlations are larger than 0.6, but at night coefficients even turn negative. In the weekend the pattern is dispersed all day long. This finding implies that annual LUR models are not sufficient to capture intra-day variation in BC concentrations.

4.2.3.2 Annual LUR model for BC

TABLE 18 shows the intercept and coefficients of the LUR model for BC based on annual average concentrations at 63 monitoring sites. R^2 of the model is 0.77. The RMSE (356.84 ng/m³) is approximately half the standard deviation (714.68 ng/m³) of the monitoring data, comparable to results presented by Mölter and others (2010b). Only traffic variables enter this model. Heavy duty traffic is influential in small buffers; total road length determines BC concentration in buffers with radius 1km. Variance inflation factors (VIF) are well below 3 indicating only limited collinearity (Eeftens et al., 2012; Johnson et al., 2010). Residuals show no spatial autocorrelation (Moran's I = -0.06; p=0.53) (Hoek et al., 2008; Ross et al., 2007). The model is validated using leave-one-out cross-validation (Brauer et al., 2003; Hoek et al., 2008); this results in a cross-validation R^2 of 0.72 and RMSE of 374 ng/m³ (appendix FIGURE A26).

TABLE 18: Annual LUR model for BC [ng/m³] (R^2=0.77; Adj R^2=0.75; RMSE=356.84; highest Cook's D=2.58)

Variable[a]	Estimate	StdErr	tValue	Probt	VIF[b]	Variable range
Intercept	811.67	150.13	5.41	0.00*	-	
ROADLENGTH_1000	0.03	0.00	10.28	0.00*	1.024	9809 – 64,003
TRAFLOADHV_FRACTION_100	4569.04	1286.25	3.55	0.00*	1.327	0 – 0.36
TRAFLOAD_50_HEAVY	393.99	112.98	3.49	0.00*	1.330	0 – 1
DIST_NEAR	-8.06	3.14	-2.56	0.01*	1.041	0.6 – 100.8

* significant at 0.05
[a] Total road length in a buffer with radius 1km (ROADLENGTH_1000), Fraction of traffic load that is heavy traffic in a buffer with radius 100m (TRAFLOADHV_FRACTION_100), Traffic load of heavy traffic in a buffer with radius 50m – indicator variable (TRAFLOAD_50_HEAVY), Distance to the nearest road (DIST_NEAR)
[b] Variance Inflation Factor

4.2.3.3 DM - Dummy model

In the model using dummy variables only two variables are included: total road length within 1km, and traffic load of heavy traffic in a buffer with radius 50m which is also a dummy variable. Both variables are significant predictors in the annual LUR model; the other two variables from the annual model do not meet the criterion of adding at least 1% to the adjusted R^2, although they are significant at the 5% level and meet predefined conditions concerning the direction of effect . The DM LUR model has an R^2 of 0.44, while the RMSE is 785.64 ng/m³ (TABLE 19). The RMSE is smaller than the standard deviation of the hourly measurements (1036.6 ng/m³).

TABLE 19: LUR model for BC [ng/m³] with 'hour' as a dummy variable (R^2=0.435; Adj R^2=0.426; RMSE=785.64; Cook's D<1 for all observations) – DM

Variable[a]	Estimate	StdErr	tValue	Probt	VIF[b]	Variable range
Intercept	527.34	106.99	4.93	0.00*	-	
ROADLENGTH_1000	0.02	0.00	23.42	0.00*	1.008	9809 – 64003
TRAFLOAD_50_HEAVY	447.04	31.30	14.28	0.00*	1.008	0 – 1
WKDY1	-257.92	139.98	-1.84	0.07	1.958	0 – 1
WKDY2	-457.85	139.98	-3.27	0.00*	1.958	0 – 1
WKDY3	-483.21	139.98	-3.45	0.00*	1.958	0 – 1
WKDY4	-454.27	139.98	-3.25	0.00*	1.958	0 – 1
WKDY5	-186.01	139.98	-1.33	0.18	1.958	0 – 1
WKDY6	355.79	139.98	2.54	0.01*	1.958	0 – 1
WKDY7	892.36	139.98	6.37	0.00*	1.958	0 – 1
WKDY8	1267.74	139.98	9.06	0.00*	1.958	0 – 1
WKDY9	749.18	139.98	5.35	0.00*	1.958	0 – 1
WKDY10	554.41	139.98	3.96	0.00*	1.958	0 – 1
WKDY11	461.49	139.98	3.30	0.00*	1.958	0 – 1
WKDY12	365.14	139.98	2.61	0.01*	1.958	0 – 1
WKDY13	246.89	139.98	1.76	0.08	1.958	0 – 1
WKDY14	359.80	139.98	2.57	0.01*	1.958	0 – 1
WKDY15	738.84	139.98	5.28	0.00*	1.958	0 – 1
WKDY16	1047.44	139.98	7.48	0.00*	1.958	0 – 1
WKDY17	1473.00	139.98	10.52	0.00*	1.958	0 – 1
WKDY18	1439.28	139.98	10.28	0.00*	1.958	0 – 1
WKDY19	1347.90	139.98	9.63	0.00*	1.958	0 – 1
WKDY20	1056.67	139.98	7.55	0.00*	1.958	0 – 1
WKDY21	887.31	139.98	6.34	0.00*	1.958	0 – 1
WKDY22	799.27	139.98	5.71	0.00*	1.958	0 – 1
WKDY23	469.90	139.98	3.36	0.00*	1.958	0 – 1
WKND0	542.63	140.54	3.86	0.00*	1.943	0 – 1
WKND1	370.92	140.54	2.64	0.01*	1.943	0 – 1
WKND2	-28.91	140.54	-0.21	0.84	1.943	0 – 1
WKND3	-234.19	140.54	-1.67	0.10	1.943	0 – 1
WKND4	-307.28	140.54	-2.19	0.03*	1.943	0 – 1
WKND5	-212.78	140.54	-1.51	0.13	1.943	0 – 1
WKND6	-85.61	140.54	-0.61	0.54	1.943	0 – 1
WKND7	-137.58	140.54	-0.98	0.33	1.943	0 – 1
WKND8	-176.82	140.54	-1.26	0.21	1.943	0 – 1
WKND9	54.17	140.54	0.39	0.70	1.943	0 – 1
WKND10	-85.63	140.54	-0.61	0.54	1.943	0 – 1
WKND11	-834.82	140.54	-5.94	0.00*	1.943	0 – 1
WKND12	-409.00	140.54	-2.91	0.00*	1.943	0 – 1
WKND13	-90.11	140.54	-0.64	0.52	1.943	0 – 1
WKND14	-77.28	140.54	-0.55	0.58	1.943	0 – 1
WKND15	-550.33	140.54	-3.92	0.00*	1.943	0 – 1

WKND16	184.78	140.54	1.31	0.19	1.943	0 – 1
WKND17	345.73	140.54	2.46	0.01*	1.943	0 – 1
WKND18	107.39	139.98	0.77	0.44	1.958	0 – 1
WKND19	169.75	139.98	1.21	0.23	1.958	0 – 1
WKND20	203.32	139.98	1.45	0.15	1.958	0 – 1
WKND21	243.87	139.98	1.74	0.08	1.958	0 – 1
WKND22	208.17	139.98	1.49	0.14	1.958	0 – 1
WKND23	28.06	140.54	0.20	0.84	1.943	0 – 1

* significant at 0.05
[a] Total road length in a buffer with radius 1km (ROADLENGTH_1000), Traffic load of heavy traffic in a buffer with radius 50m – indicator variable (TRAFLOAD_50_HEAVY), weekday hour 0-23 (WKDYx), weekend hour 0-23 (WKNDx)
[b] Variance Inflation Factor

4.2.3.4 HM1 – Hourly model 1

In TABLE 20 (weekday) and in the appendix TABLE A11 (weekend day) an overview is given of 48 hourly LUR models always using the same predictors. R^2 values range from 0.05 to 0.73; RMSE ranges from 321 ng/m³ to 1567 ng/m³. The R^2 of hourly models is always smaller than the R^2 of the annual model, although between 6 a.m. and 10 p.m. on weekdays the R^2 approximates the annual model R^2. Variables that are significant in the annual LUR model are not necessarily significant in an hourly LUR model. The direction of effect of parameters in the hourly model sometimes changes, in a way that is inconsistent with a priori defined conditions. Models for night hours have lower R^2 values caused by the smaller range in concentrations between different locations.

4.2.3.5 HM2 – Hourly model 2

Because significant variables are selected per hour, this leads to higher hourly R^2 values (0.07 – 0.8) and lower RMSE (287 ng/m³ - 1407 ng/m³) compared to HM1 (TABLE 21 (weekday) and appendix TABLE A12 (weekend day)). Between 2 p.m. and 4 p.m. on weekdays individual hourly models perform better than the annual model for the R^2, however the RMSE is larger. During the day traffic variables (heavy traffic and distance to the nearest (major) road) often enter the model. Traffic intensity (light traffic only) is significant on peak hours (8 a.m., 9 a.m., 5 p.m., 7 p.m.). On weekday nights, land use variables in larger buffers come into the model, but the limited concentration contrast results in low R^2, and the concentrations are mainly predicted by the intercept. On weekend nights BC concentrations were elevated and traffic variables are significant, indicative of busy traffic on weekend nights.

4.2.3.6 HM3 – Hourly model 3

Results of the hourly LUR models using dynamic covariates are presented in TABLE 22 (weekday) and in the appendix TABLE A13 (weekend day). Dynamic variables that enter the models are indicated in bold. The selected covariates and the models' performance are very similar to those of the previous models (TABLE 21). Adding the dynamic complement of the static variables does not really improve, and sometimes even deteriorated the hourly LUR models. On weekdays at 4 a.m. traffic intensity on the nearest major road was included in the LUR model, but in the model with dynamic variables this variable turned out to be not predictive. On weekdays at 9 p.m. and 10 p.m. population density was removed from the model and other variables became significant and improved the model. Since population density was the first variable to enter the model, it interacted with the other significant variables. Because the dynamic complement of the population variable was not significant anymore, other variables or other groups of variables became significant and even improved the model. This illustrates that the forward stepwise regression procedure does not automatically select the best model. A lagged effect of dynamic variables was considered, but hourly variables are highly correlated (e.g. correlation between People_1000 on different hours is always > 0.99; correlation between QD_near1 on different hours is always > 0.95), making it very hard to detect a real effect. When selecting for example traffic on weekdays at 8 a.m. and traffic at 7 a.m. in one model, this leads to very high VIFs.

TABLE 20: Hourly LUR models for BC [ng/m³] while keeping the significant variables of the annual LUR model (weekday hours) – HM1

						Highest		ROADLENGTH_1000 [a]		TRAFLOADHV_FRACTION_100 [a]		TRAFLOAD_50_HEAVY [a]		DIST_NEAR [a]	
Day	hour	N	R^2	Adj R^2	RMSE	Cook's D	β_0	β_1	prob1	β_2	prob2	β_3	prob3	β_4	prob4
WKDY	0	63	0.14	0.08	451.18	<1	1155	0.01	0.01	-2129.02	0.20	163.05	0.26	1.03	0.80
WKDY	1	63	0.09	0.03	351.81	1.11	1141	0.00	0.15	-2161.40	0.09	75.81	0.50	2.84	0.36
WKDY	2	63	0.11	0.05	327.38	<1	1290	0.00	0.43	-2621.30	0.03	16.61	0.87	-0.10	0.97
WKDY	3	63	0.10	0.04	390.09	<1	1420	-0.01	0.13	-1904.14	0.18	-65.20	0.60	-1.74	0.62
WKDY	4	63	0.05	-0.02	320.60	<1	1192	0.00	0.82	-1797.70	0.13	132.48	0.20	-1.24	0.66
WKDY	5	63	0.21	0.15	367.35	2.36	1198	0.00	0.29	2695.68	0.05	175.15	0.14	-1.57	0.63
WKDY	6	63	0.46	0.43	432.62	1.50	1196	0.01	0.00	3027.30	0.06	472.37	0.00	-1.80	0.64
WKDY	7	63	0.53	0.50	607.65	<1	1243	0.03	0.00	4568.74	0.04	575.40	0.00	-8.92	0.10
WKDY	8	63	0.43	0.39	962.82	<1	1392	0.03	0.00	6873.58	0.05	562.59	0.07	-17.06	0.05
WKDY	9	63	0.55	0.52	604.92	1.90	1002	0.03	0.00	5715.53	0.01	336.62	0.08	-10.61	0.05
WKDY	10	63	0.46	0.42	653.51	2.44	892	0.03	0.00	5491.59	0.02	163.44	0.43	-8.84	0.13
WKDY	11	63	0.64	0.62	585.29	1.07	281	0.04	0.00	4999.53	0.02	562.17	0.00	-6.35	0.22
WKDY	12	63	0.59	0.57	661.00	1.96	230	0.04	0.00	6267.88	0.01	550.19	0.01	-7.46	0.21
WKDY	13	63	0.60	0.57	625.99	3.18	145	0.04	0.00	3002.61	0.19	763.64	0.00	-5.41	0.33
WKDY	14	63	0.69	0.66	511.55	1.88	266	0.04	0.00	2577.89	0.17	601.31	0.00	-8.81	0.06
WKDY	15	63	0.73	0.71	606.42	1.86	96	0.05	0.00	4811.63	0.03	776.25	0.00	-8.69	0.11
WKDY	16	63	0.72	0.70	679.66	3.27	130	0.06	0.00	3986.39	0.11	907.84	0.00	-10.32	0.09
WKDY	17	63	0.67	0.65	831.74	2.18	518	0.06	0.00	-800.48	0.79	1092.14	0.00	-19.50	0.01
WKDY	18	63	0.60	0.57	883.51	1.63	706	0.06	0.00	-2146.18	0.50	962.51	0.00	-17.47	0.03
WKDY	19	63	0.58	0.55	759.48	<1	935	0.05	0.00	-3163.64	0.25	749.10	0.00	-12.20	0.07
WKDY	20	63	0.57	0.54	580.46	<1	1053	0.04	0.00	-1622.53	0.44	526.89	0.01	-5.75	0.27
WKDY	21	63	0.49	0.46	551.40	<1	1242	0.03	0.00	-2246.63	0.26	544.82	0.00	-5.48	0.26
WKDY	22	63	0.40	0.36	575.80	<1	1241	0.03	0.00	-785.90	0.71	320.55	0.08	-3.98	0.44
WKDY	23	63	0.20	0.15	603.70	<1	1473	0.02	0.00	-2832.36	0.20	84.08	0.66	-6.05	0.26
Year		63	0.77	0.75	356.84	2.58	812	0.03	0.00	4569.04	0.00	393.99	0.00	-8.06	0.01

[a] Total road length in a buffer with radius 1km (ROADLENGTH_1000), Fraction of traffic load that is heavy traffic in a buffer with radius 100m (TRAFLOADHV_FRACTION_100), Traffic load of heavy traffic in a buffer with radius 50m – indicator variable (TRAFLOAD_50_HEAVY), Distance to the nearest road (DIST_NEAR)

TABLE 21: Hourly LUR models for BC [ng/m³] using hourly monitoring data and static independent variables: independent models (weekday hours) – HM2

hour	N	R^2	Adj R^2	RMSE	Highest Cook's D	β_0	β_1	X_1 [a]	β_2	X_2 [a]	β_3	X_3 [a]	β_4	X_4 [a]	β_5	X_5 [a]	β_6	X_6 [a]
0	63	0.24	0.22	415.89	<1	1601	2.6×10^{-4}	HDRES_1000	-4.5×10^{-2}	D_HIGH								
1	63	0.18	0.17	325.04	<1	1264	2.3×10^{-4}	HDRES_1000										
2	63	0.16	0.13	311.92	<1	1335	-7.4×10^{-5}	URBGR_3000	-200	URBGR_500								
3	63	0.15	0.14	369.00	<1	1366	-1.3×10^{-4}	URBGR_3000										
4	63	0.23	0.19	286.84	<1	1412	-1.6×10^{-4}	URBGR_3000	-7.0×10^{-4}	NATURE_500	6.3×10^{-3}	Q_NEAR_MAJOR						
5	63	0.30	0.27	342.01	<1	1164	4409	TRAFLOADHV_FRACTION_100	289	HDRES_500	1.2×10^{-4}	IND_3000						
6	63	0.54	0.51	399.21	1.34	1761	451	TRAFLOAD_50_HEAVY	-14.1×10^{-2}	DIST_NEAR_MAJOR	2.8×10^{-4}	HDRES_1000	3560	TRAFLOADHV_FRACTION_100				
7	63	0.59	0.56	573.65	<1	967	1.7×10^{-2}	ROADLENGTH_1000	688	TRAFLOAD_50_HEAVY	2.7×10^{-2}	TRAFLOADHV_FRACTION_100_2	5.2	ADDRESS_100	1.8×10^{-4}	IND_3000		
8	63	0.60	0.57	814.40	<1	1293	40.9×10^{-2}	ADDRESS_1000	3.5×10^{-2}	TRAFLOADHV_FRACTION_100_2	-2.2×10^{-4}	URBGR_5000	6.9	QD_NEAR1_HEAVY	2.4×10^{-2}	Q_NEAR_MAJOR		
9	63	0.64	0.61	543.37	<1	1268	27.8×10^{-2}	ADDRESS_1000	3.7×10^{-2}	TRAFLOADHV_FRACTION_100_2	-1.0×10^{-4}	URBGR_5000	1.5×10^{-2}	Q_NEAR_MAJOR	-9.7	DIST_NEAR		
10	63	0.54	0.52	595.30	<1	1270	12.0×10^{-2}	ADDRESS_1000	8665	TRAFLOADHV_FRACTION_100	4.3×10^{-4}	HDRES_1000						
11	63	0.75	0.73	492.70	<1	379	4.3×10^{-2}	ROADLENGTH_1000	13454	D_NEAR_MAJOR1	8655	TRAFLOADHV_FRACTION_100	-2.2×10^{-4}	URBGR_3000				
12	63	0.68	0.65	589.63	1.67	336	4.3×10^{-2}	ROADLENGTH_1000	4.0×10^{-2}	TRAFLOADHV_FRACTION_100_2	12015	D_NEAR_MAJOR1	-2.4×10^{-4}	URBGR_3000				
13	63	0.74	0.72	504.07	<1	838	17223	D_NEAR_MAJOR1	23.4×10^{-2}	ADDRESS_1000	8720	TRAFLOADHV_FRACTION_100	-2.3×10^{-4}	URBGR_3000				
14	63	0.79	0.76	428.76	1.55	504	3.4×10^{-2}	ROADLENGTH_1000	10316	D_NEAR_MAJOR1	339	TRAFLOAD_50_HEAVY	-8.3	DIST_NEAR	4236	TRAFLOADHV_FRACTION_100	-283	URBGR_1000
15	63	0.80	0.78	524.30	1.87	78	4.2×10^{-2}	ROADLENGTH_1000	417	TRAFLOAD_50_HEAVY	14083	D_NEAR_MAJOR1	6354	TRAFLOADHV_FRACTION_100	592	D_NEAR1		
16	63	0.79	0.77	602.37	2.55	712	2.7×10^{-2}	ROADLENGTH_1000	612	TRAFLOAD_50_HEAVY	11486	D_NEAR_MAJOR1	5003	TRAFLOADHV_FRACTION_100	5.1×10^{-2}	PEOPLE_1000	-11.3	DIST_NEAR
17	63	0.72	0.70	772.92	<1	941	3.4×10^{-2}	ROADLENGTH_1000	797	TRAFLOAD_50_HEAVY	-17.9	DIST_NEAR	32.5×10^{-2}	QD_NEAR1	8.2×10^{-5}	HDRES_5000		

18	63	0.63	0.61	844.24	<1	1741	11.4×10^{-2}	PEOPLE_1000	888	TRAFLOAD_50_HEAVY	-18.6	DIST_NEAR		
19	63	0.59	0.57	737.64	1.05	1708	9.5×10^{-2}	PEOPLE_1000	45.2×10^{-2}	QD_NEAR1				
20	63	0.55	0.54	579.17	<1	927	3.8×10^{-2}	ROADLENGTH_1000	480	TRAFLOAD_50_HEAVY				
21	63	0.49	0.47	543.38	<1	1669	5.9×10^{-2}	PEOPLE_1000	483	TRAFLOAD_50_HEAVY				
22	63	0.42	0.41	555.35	<1	1661	5.7×10^{-2}	PEOPLE_1000	313	TRAFLOAD_50_HEAVY				
23	63	0.33	0.30	545.80	3.10	2015	4.6×10^{-4}	HDRES_1000	-5.6×10^{-2}	D_HIGH				
Year	63	0.77	0.75	356.84	2.58	812	3.1×10^{-2}	ROADLENGTH_1000	4569	TRAFLOADHV_FRACTION_100	394	TRAFLOAD_50_HEAVY	-8.1	DIST_NEAR

[a] High density residential in a buffer with size XXm (HDRES_XX), Distance to the nearest highway (D_HIGH), Urban green in a buffer with size XXm (URBGR_XX), Natural land in a buffer with size XXm (NATURE_XX), Traffic intensity (private cars) on the nearest major road (Q_NEAR_MAJOR), Fraction of heavy traffic in a buffer with size XXm (TRAFLOADHV_FRACTION_XX), Industrial or commercial units in a buffer with size XXm (IND_XX), Sum of (traffic intensity (heavy traffic) * road length) in a buffer with size XXm (TRAFLOAD_XX_HEAVY), Distance to the nearest major road (DIST_NEAR_MAJOR), Total road length in a buffer with size XXm (ROADLENGTH_XX), Fraction of heavy traffic squared in a buffer with size XXm (TRAFLOADHV_FRACTION_XX_2), Number of addresses in a buffer with size XXm (ADDRESS_XX), Traffic intensity (heavy traffic) on the nearest road / distance to the nearest road (QD_NEAR1_HEAVY), Distance to the nearest road (DIST_NEAR), 1 / Distance to the nearest major road (D_NEAR_MAJOR1), Number of people in a buffer with size XXm (PEOPLE_XX), Traffic intensity (private cars) on the nearest road / distance to the nearest road (QD_NEAR1)

TABLE 22: Hourly LUR models for BC [ng/m³] using hourly monitoring data and dynamic independent variables: independent models (weekday hours) – HM3

hour	N	R^2	Adj R^2	RMSE	Highest Cook's D	β_0	β_1	X_1 [a]	β_2	X_2 [a]	β_3	X_3 [a]	β_4	X_4 [a]	β_5	X_5 [a]	β_6	X_6 [a]
0	63	0.24	0.22	415.89	<1	1601	2.6×10^{-4}	HDRES_1000	-4.5×10^{-2}	D_HIGH								
1	63	0.18	0.17	325.04	<1	1264	2.3×10^{-4}	HDRES_1000										
2	63	0.16	0.13	311.92	<1	1335	-7.4×10^{-5}	UrbGr_3000	-200	UrbGr_500								
3	63	0.15	0.14	369.00	<1	1366	-1.3×10^{-4}	UrbGr_3000										
4	63	0.16	0.14	295.51	<1	1504	-1.5×10^{-4}	UrbGr_3000	-6.5×10^{-4}	NATURE_500								
5	63	0.30	0.27	342.01	<1	1164	4409	TRAFLOADHV_FRACTION_100	289	HDRES_500	1.2×10^{-4}	IND_3000						
6	63	0.54	0.51	399.21	1.34	1761	451	TRAFLOAD_50_HEAVY	-14.1×10^{-2}	DIST_NEAR_MAJOR	2.8×10^{-4}	HDRES_1000	3560	TRAFLOADHV_FRACTION_100				
7	63	0.59	0.56	573.65	<1	967	1.7×10^{-2}	ROADLENGTH_1000	688	TRAFLOAD_50_HEAVY	2.7×10^{-2}	TRAFLOADHV_FRACTION_100_2	5.2	ADDRESS_100	1.8×10^{-4}	IND_3000		
8	63	0.58	0.54	836.08	<1	1376	39.9×10^{-2}	ADDRESS_1000	3.5×10^{-2}	TRAFLOADHV_FRACTION_100_2	-2.2×10^{-4}	URBGR_5000	6.8	QD_NEAR1_HEAVY	29.9×10^{-2}	**Q_NEAR_MAJOR_WKDY8**		
9	63	0.64	0.61	543.40	<1	1270	27.9×10^{-2}	ADDRESS_1000	3.6×10^{-2}	TRAFLOADHV_FRACTION_100_2	-1.0×10^{-4}	URBGR_5000	25.8×10^{-2}	**Q_NEAR_MAJOR_WKDY9**	-9.9	DIST_NEAR		
10	63	0.54	0.52	595.30	<1	1270	12.0×10^{-2}	ADDRESS_1000	8665	TRAFLOADHV_FRACTION_100	4.3×10^{-4}	HDRES_1000						
11	63	0.75	0.73	492.70	<1	379	4.3×10^{-2}	ROADLENGTH_1000	13454	D_NEAR_MAJOR1	8655	TRAFLOADHV_FRACTION_100	-2.2×10^{-4}	URBGR_3000				
12	63	0.68	0.65	589.63	1.67	336	4.3×10^{-2}	ROADLENGTH_1000	4.0×10^{-2}	TRAFLOADHV_FRACTION_100_2	12015	D_NEAR_MAJOR1	-2.4×10^{-4}	URBGR_3000				
13	63	0.76	0.74	490.92	1.59	789	8.0×10^{-2}	**PEOPLE_1000_WKDY13**	368	TRAFLOAD_50_HEAVY	13083	D_NEAR_MAJOR1	19.7×10^{-4}	IND_3000	6470	TRAFLOADHV_FRACTION_100	-2.1×10^{-4}	URBGR_3000
14	63	0.79	0.76	428.76	1.55	504	3.4×10^{-2}	ROADLENGTH_1000	10316	D_NEAR_MAJOR1	339	TRAFLOAD_50_HEAVY	-8.3	DIST_NEAR	4236	TRAFLOADHV_FRACTION_100	-283	URBGR_1000
15	63	0.80	0.78	520.77	<1	137	4.2×10^{-2}	ROADLENGTH_1000	1546	**TRAFLOADMAJOR_50_WKDY15**	8338	TRAFLOADHV_FRACTION_100	672	D_NEAR1				
16	63	0.79	0.77	604.58	2.62	698	2.8×10^{-2}	ROADLENGTH_1000	609	TRAFLOAD_50_HEAVY	11420	D_NEAR_MAJOR1	4958	TRAFLOADHV_FRACTION_100	4.8×10^{-2}	**PEOPLE_1000_WKDY16**	-11.4	DIST_NEAR
17	63	0.73	0.70	769.86	<1	970	3.3×10^{-2}	ROADLENGTH_1000	773	TRAFLOAD_50_HEAVY	-18.0	DIST_NEAR	3.5	**QD_NEAR1_WKDY17**	0.8×10^{-4}	HDRES_5000		

18	63	0.65	0.63	821.32	<1	1775	10.9×10^{-2}	**PEOPLE_1000_WKDY18**	609	TRAFLOAD_50_HEAVY	-17.8	DIST_NEAR	3.8	**QD_NEAR1_WKDY18**
19	63	0.62	0.60	716.14	<1	1937	9.3×10^{-2}	**PEOPLE_1000_WKDY19**	7.2	**QD_NEAR1_WKDY19**	-13.0	DIST_NEAR		
20	63	0.55	0.54	579.17	<1	927	3.8×10^{-2}	ROADLENGTH_1000	480	TRAFLOAD_50_HEAVY				
21	63	0.50	0.48	538.87	<1	1528	17.3×10^{-2}	ADDRESS_1000	508	TRAFLOAD_50_HEAVY				
22	63	0.43	0.41	550.86	<1	1822	1.3×10^{-4}	HDRES_3000	1.6×10^{-3}	TRAFLOAD_100_HEAVY				
23	63	0.33	0.30	545.80	3.10	2015	4.6×10^{-4}	HDRES_1000	-5.6×10^{-2}	D_HIGH				
Year	63	0.77	0.75	356.84	2.58	812	3.1×10^{-2}	ROADLENGTH_1000	4569	TRAFLOADHV_FRACTION_100	394	TRAFLOAD_50_HEAVY	-8.1	DIST_NEAR

Dynamic variables that enter the model are indicated in bold.

[a] High density residential in a buffer with size XXm (HDRES_XX), Distance to the nearest highway (D_HIGH), Urban green in a buffer with size XXm (URBGR_XX), Natural land in a buffer with size XXm (NATURE_XX), Fraction of heavy traffic in a buffer with size XXm (TRAFLOADHV_FRACTION_XX), Industrial or commercial units in a buffer with size XXm (IND_XX), Sum of (traffic intensity (heavy traffic) * road length) in a buffer with size XXm (TRAFLOAD_XX_HEAVY), Distance to the nearest major road (DIST_NEAR_MAJOR), Total road length in a buffer with size XXm (ROADLENGTH_XX), Fraction of heavy traffic squared in a buffer with size XXm (TRAFLOADHV_FRACTION_XX_2), Number of addresses in a buffer with size XXm (ADDRESS_XX), Traffic intensity (heavy traffic) on the nearest road / distance to the nearest road (QD_NEAR1_HEAVY), Traffic intensity (private cars) on the nearest major road at hour x (Q_NEAR_MAJOR_WKDYx), Distance to the nearest road (DIST_NEAR), 1 / Distance to the nearest major road (D_NEAR_MAJOR1), Number of people in a buffer with size XXm at hour x (PEOPLE_XX_WKDYx), Sum of (traffic intensity on major roads (private cars) at hour x * road length of major roads) in a buffer with size XXm (TRAFLOADMAJOR_XX_WKDYx), 1 / Distance to the nearest road (D_NEAR1), Traffic intensity (private cars) on the nearest road at hour x / distance to the nearest road (QD_NEAR1_WKDYx)

4.2.4 DISCUSSION

LUR models for the carbonaceous component of particulate matter, BC, were developed. Due to the difficulty of measuring BC (labor intensive and expensive measurement techniques), only very few LUR models for BC exist (Brauer et al., 2008; Gryparis et al., 2007). Reflectance of PM filters is often used more or less synonymously of BC. Models for $PM_{2.5}$ absorbance ($PM_{2.5}$abs) are more widespread and regularly made together with models for PM_{10} and $PM_{2.5}$ (Clougherty et al., 2008; Eeftens et al., 2012; Henderson et al., 2007; Morgenstern et al., 2007). The R^2 of the annual model in this paper (R^2=0.77) is similar to explained variances in other study areas. The number of measurement sites is larger than in previous LUR models for BC or $PM_{2.5}$abs (63 compared to ~40 sites), and that probably results in slightly lower R^2 values (Basagaña et al., 2012; Wang et al., 2012), although the model developed here will be better when applying it to a hold-out sample. The study is restricted by the rather short sampling period. On most locations, only 2 weeks of measurements were available and that might limit the validity of the resulting models. Because measurements originated from two different campaigns, the first one in 2010 and the second one in 2011, comparability of annual average concentrations was a necessary precondition for joining both datasets.

Hourly LUR models for BC are developed using different strategies: by means of dummy variables, with time-resolved dependent variables and/or with dynamic independent variables. Low Spearman coefficients (Avg=0.38; SD=0.26) suggested that one annual LUR model was not sufficient to capture variations in BC concentrations during the day. This is in contrast to other studies defining LUR models on different time scales (e.g. season): in those study areas it was demonstrated that spatial variability in NO_2 concentrations remained consistent across the sampling periods (Beelen et al., 2007; Crouse et al., 2009; Wheeler et al., 2008). Covariates that often proved significant were traffic variables (heavy traffic in smaller buffers, total road length within 1km and distance to the nearest (major) road), population variables (number of addresses or number of people in a buffer with radius 1km), and land use variables (residential land use, nature or urban green, in buffers > 1km). Light and heavy traffic originate from a different dataset because for light traffic we had the opportunity to use hourly traffic streams. Yet light and heavy traffic streams were highly correlated and heavy traffic variables were often excluded before the regression model was estimated. The fraction of heavy traffic was less correlated to other traffic variables, and seemed to replace the absolute number of passing trucks in the models. Moreover by using the fraction, it was assumed that not only the number of trucks, but also the environment (highway versus urban road) influences air quality; e.g. local urban roads with a high fraction of heavy traffic but a low number of trucks might still cause an important impact. Distance to the nearest road has been used as a predictor variable in many published LUR studies (Brauer et al., 2008; Hochadel et al., 2006; Mölter et al., 2010a). In this study, it was offered to the model in a linear form and in an exponential form, and both also in combination with traffic intensity. Linear distance was only included in the model when a traffic intensity variable (light or heavy traffic) in a small buffer was also included. In the hourly LUR models with significant variables of the annual model (HM1), distance to the nearest road was only significant on traffic peak hours and the coefficient was more influential on those peak hours, illustrating the relationship between distance and traffic intensity. A model where distance to the nearest road is included, can be applied solely to predict concentrations at the facade of houses, whereas at larger distances concentrations can become negative. Traffic volume of all traffic is expected to be significant in several LUR models, especially on traffic peak hours and in smaller buffers (Kim and Guldmann, 2011). The insignificance of traffic intensity variables for light traffic in the annual model is caused by the sparseness of the road network used to assign traffic streams from the FEATHERS model: too many locations have no predicted traffic intensities on the nearest road. Forcing traffic volume on the nearest road into the annual model (variable is not significant at 0.05) increases the R^2 from 0.7668 to 0.7745, but the new variable only contributes between 4 ng/m³ and 441 ng/m³ to the modeled concentrations (on average 32 ng/m³). Meteorological variables were not collected, although by rescaling the measurements based on hourly variations in background concentrations, meteorological effects near the reference monitor are taken into account.

During model development, we encountered high Cook's D values in some models, pointing to influential observations. Removing the observation that substantially influences the model is one option, although in this study we cannot motivate this decision. Another option is to exclude a variable from model development: remove the variable that is not significant anymore when holding out the influential observation (Eeftens et al., 2012). In our case, the variable that influences the model, often fraction of heavy traffic, also returns in other hourly models without a high Cook's D value for the same observations; this suggests that the variable is not accidentally included. Besides when building an annual LUR model based on the urban measurement campaign alone (42 mostly urban locations; R^2=0.73 and RMSE=290 for model with an influential observation; R^2=0.68 and RMSE=311 for model without influential observation; appendix FIGURE A27), and applying the model to the hold-out regional sample (21 urban and regional locations), the model with a high Cook's D value was much better in predicting concentrations compared to the reformed model with all Cook's D's < 1 (validation R^2=0.50, validation RMSE=627.69; compared to validation R^2=0.21, validation RMSE=822.51). Therefore it was decided not to correct for high Cook's D values in this sample.

Temporal variation from a continuous background monitoring station can be applied to an existing annual LUR model (similar to Gan and others (2011), Nethery and others (2008b), and Slama and others (2007)), but in this case the temporal trend of only one station is implemented to all other locations in the study area. This method is easy and effective when one is interested in concentrations on a specific date, although ignoring the intra-day variation in concentrations on different types of locations (Wang et al., 2013). Retrieving hourly values with this method would inherently lead to large uncertainties; e.g. if a local event impacts the concentrations near the background monitor, predicted concentrations on all other locations would be affected. Another straightforward method to add a temporal trend to a LUR model, would be to simply replace a variable with its dynamic complement, at least if in the annual model a population density variable or traffic intensity variable (light traffic) is inserted (Chen et al., 2010).

Comparing between different methodologies to develop hourly LUR models is difficult. With 'hour' as a dummy variable, one LUR model is developed, with one set of model performance criteria. In the other methodologies hourly models are constructed with hourly performance criteria. The R^2 of the dummy model (0.44) is remarkably lower than the R^2 of the annual model (0.77), but this is still acceptable taking into account that the number of observations is multiplied by 48. The same spatial pattern is assumed all day long in the dummy model, which conflicts with our findings based on the hourly measurements of BC. Moreover hourly concentrations are not independent of each other, violating the assumption of independence in linear regression. Keeping significant variables of the annual LUR model and estimating coefficients again with hourly observations (HM1), results in several insignificant variables, and the variables often do not meet a priori defined conditions with respect to direction of effect. Building new hourly LUR models independent of each other (HM2) solves the issue of insignificant variables or variables with a wrong direction of effect. Even though the hourly LUR models are developed independently of adjacent hours, in fact similar variables often return in consecutive models demonstrating the robustness of the models. This also suggests that hourly models could be combined and that less than 24 unique models are necessary to capture intra-day variation in concentrations. Grouping of hours could be based on the correlation between measurements on different hours, or it could be based on the similarity of the developed hourly LUR models: this should be explored further. Adding dynamic predictor variables instead of static variables (HM3) did not drastically improve the hourly LUR models, and sometimes even deteriorated the models. Lagged effects were considered, but a major problem is the high correlation between the hourly dynamic variables. For BC, a lagged effect is probably not present because BC from traffic has a very local effect and concentrations tend to decrease very fast when moving away from the source. For secondary pollutants like NO_2, including lagged effects could be more promising and worth considering in future work.

An important advantage of LUR models is the good cost-benefit ratio compared to dispersion models; but building 48 single LUR models might undermine this point. In our experience, hourly LUR models are not more data-demanding than annual LUR models, except for the models with dynamic covariates (HM3).

Time-resolved measurements should be available, but the use of an active monitor does not necessarily require more supervision than passive samplers once in place. Estimating 48 LUR models obviously takes more time than developing one annual LUR model, but model development can be largely automated and running the script takes only seconds, even on a laptop computer.

The hourly LUR models developed in this study can be considered as annual average models with an hourly temporal resolution. Seasonal variability is not accounted for as this variability is mostly driven by meteorological conditions and wood smoke, whereas diurnal variability is principally derived from human activities. Covariates explaining diurnal variability, e.g. traffic flows, are included as covariates in the models. Hourly LUR models are useful in determining long or medium term personal exposure to air pollution more accurately when combined with time-activity data. Agent-based models, like activity-based traffic models, are capable of predicting those diaries; in the future the whereabouts will be linked to the hourly LUR models to predict exposure of modeled agents (Int Panis, 2010). For long term epidemiological studies, the added value of using hourly LUR models should be investigated, although it is clear that this method will be able to capture personal exposure in more detail. Alternatively, hourly LUR models can also improve epidemiological studies focusing on acute health effects. Diaries or GPS loggers can be deployed in large quantities to register the movement of individuals (de Nazelle et al., 2013; Dons et al., 2013a) and hourly LUR models can then be used to estimate personal exposure.

4.3 IMPLEMENTATION AND VALIDATION OF A MODELING FRAMEWORK TO ASSESS PERSONAL EXPOSURE TO BLACK CARBON

This chapter is based on:

Dons, E., Van Poppel, M., Kochan, B., Wets, G., Int Panis, L., 2013. Implementation and validation of a modeling framework to assess personal exposure to black carbon. Submitted to Environment International.

4.3.1 INTRODUCTION

High-resolution personal measurements are the best way of getting information on the exposure of individuals; which combined with geo-information enables exposure to be assessed in space and time. Unfortunately personal measurements are expensive and a burden to individuals; therefore most current studies are limited to short periods of personal measurements in a small sample. These snapshots of exposure are not sufficient to provide epidemiologists with data on long term exposure of large cohorts. Building exposure models is an obvious alternative, but this often results in simple models not capable of estimating exposure with enough reliability.

Ideally personal exposure to air pollution should be modeled as a combination of two interacting geographies: a moving population and a continuously changing air quality (Briggs, 2005). Many studies only take into account residential location and ignore time-activity patterns while air pollution is typically modeled using annual average concentrations without any daily variation. Several more complex exposure models have been developed: e.g. SHEDS (Stochastic Human Exposure and Dose Simulation Model, (Burke et al., 2001)), pCNEM (Probabilistic Version of the NAAQS Exposure Model, (Zidek et al., 2005)), STEMS (Space-Time Exposure Modeling System, (Gulliver and Briggs, 2005)), EMI (Exposure Model for Individuals, (Breen et al., 2010)), MEEM (MicroEnvironmental Exposure Model, (Mölter et al., 2012)). The first two models estimate population exposure; the other models assess exposure of individuals. Models estimating exposure of individuals are up till now not designed to model exposure of a full population as specific information on individuals, not available on an aggregated level, is required. Population exposure models generate diaries from activity databases, often without a geographical dimension, and often deterministic in nature. Beckx et al. (2009b) and Hatzopoulou et al. (2011) were the first to use an activity-based transportation model to predict stochastic time-location-activity diaries and use them in an air pollution exposure model. Lefebvre et al. (2013a) and Dhondt et al. (2012b) used activity-based models to evaluate the effect of air quality measures on population exposure and health. An important issue is that the final outcome of the aforementioned population exposure models has never been fully validated; the validation was limited to the validation of submodels. Personal monitoring of a random sample of *n* individuals from a target population is a valuable tool to examine the validity of human exposure models (Duan, 1991; Gerharz et al., 2013; Mölter et al., 2012). Models for individual exposure assessment expect diaries or GPS tracks to be known: validating these kinds of models in fact means validating only the air quality models, and not the diaries themselves. In the case of population exposure models, the space-time predictions need to be validated as well, together with the air quality models.

In this study, a personal exposure modeling framework using an activity-based model will be implemented and its performance will be compared to real-life personal exposures. Most submodels have already been presented elsewhere (Bellemans et al., 2010; Dons et al., 2013a; Dons et al., 2013b), but in this study all the models are integrated in one framework for the first time: the AB²C model (An Activity-Based modeling framework for Black Carbon exposure assessment). Personal measurements are compared against a distribution of personal exposure estimates and serve as a validation for the complete framework. The AB²C model is developed to estimate population-wide exposure to black carbon (BC), a pollutant suspected of having health effects (WHO, 2012; Zanobetti and Schwartz, 2006). Introducing individual mobility in population exposure models for BC is highly relevant because moving from one place to another can significantly alter exposure through the steep concentration gradients observed near BC sources. Individual exposure assessment on a population level, as aimed for by the AB²C model, can contribute to health policy: highly exposed individuals or groups of individuals can be identified; the impact of age, gender or socio-economic class on exposure can be verified or the number of individuals exposed to levels above a certain threshold can be determined. Moreover policy scenarios can be calculated and the effect on personal and population exposure to BC can be assessed, and ultimately extended with appropriate exposure-response functions to perform health impact assessments.

4.3.2 MATERIALS AND METHODS

The modeling framework AB²C consists of an activity-based model, hourly land use regression models, an in-traffic personal exposure model, and an indoor air model. Separate models are described first; how models fit in a consistent framework is discussed afterwards. Real-life measurements serve as a validation of the entire model chain.

4.3.2.1 Activity-based model

Activity-based models simulate activities and trips for a synthetic population based on thousands of revealed diaries (Arentze and Timmermans, 2004; Davidson et al., 2007). Trips are derived from the activities performed and their physical location: if subsequent activities are in a different geographical zone, a trip between the origin and the destination is required. In Flanders, FEATHERS (Forecasting Evolutionary Activity-Travel of Households and their Environmental RepercussionS) was developed as a simulation platform to implement activity-based models (Bellemans et al., 2010; Kochan et al., 2013). For every adult in the population, approximately 6 million people, a diary is built using a series of decision trees and constraints. It results in a model predicting activities (home-based activities, work/education, business, bring/get, shopping, services, social, leisure, touring, other), their timing, duration and location for 24h for every agent in a synthetic population. The study area is divided into 2386 subzones, also called Traffic Analysis Zones (TAZ; terminology used in transportation planning models), with an average size of 5.7 km²; the size of a subzone is related to the number of inhabitants. The subzone where an activity will be performed is determined in FEATHERS by taking into account land use data such as the number of work places and the number of inhabitants in each TAZ to estimate the level of production and attraction. Agents have a preference of performing an activity closer to home for shopping trips, but the distance can be larger for work locations, etc.. For all 168 hours of an average week, a dynamic population density is calculated, not based on static addresses, but based on the locations that agents actually visit during each hour. For trips, the transport mode (car driver, car passenger, bike/walk, public transport), the duration of the trip, and subzone of origin and destination will be predicted by FEATHERS. Those trips are then assigned to a road network using an equilibrium assignment, thus taking into account congestion effects, in TransCAD software (Caliper, 2013). Outputs from the FEATHERS model used in subsequent steps are: hourly traffic flows for motorized trips, hourly dynamic population densities taking into account population mobility, and 24h-diaries for a subset of agents with specific characteristics (see appendix TABLE A14 for an example output of a modeled diary).

4.3.2.2 Hourly LUR models

In a land use regression (LUR) model statistical associations are developed between potential predictor variables and measured pollutant concentrations as a basis for predicting concentrations at unsampled sites (Hoek et al., 2008; Ryan and LeMasters, 2007). In 2010 and 2011, BC was measured on 63 locations in Flanders with a high temporal resolution (5-min) using micro-aethalometers (AethLabs, 2011; Dons et al., 2013b). Annual average concentrations ranged from 846 ng/m³ to 4184 ng/m³, with hourly concentration peaks of nearly 10,000 ng/m³. Hourly LUR models were derived for weekdays (Mon-Fri, 24 models) and for the weekend (Sat-Sun, 24 models) to capture intraday variation in the spatial concentration pattern of the study area (chapter 4.2). Seasonal variability is not accounted for as this variability is generally driven by meteorological conditions and meteorological variables are not collected, whereas diurnal variability is principally derived from human activities. The hourly models were estimated independent of each other according to a fixed model development algorithm (Eeftens et al., 2012; Henderson et al., 2007). Weekday hourly models performed well during the day and on traffic peak hours, explaining 60 to 80% of variability using mainly traffic variables (chapter 4.2). At night and in the weekend the models were less predictive using only 1 or 2 predictors, but RMSE values were low indicative of homogeneous BC concentrations (chapter 4.2). Traffic and population variables from the activity-based model were only sporadically included.

In this study, concentrations were calculated for 10 randomly chosen buildings in each subzone using the hourly LUR models; the median concentration is the exposure assigned to people present in that subzone during that hour (10 buildings proved to be sufficient to reliably estimate median concentrations, see appendix A.4 for details on the methodology).

4.3.2.3 In-traffic personal exposure model

While exposure in geographical places can be modeled using hourly LUR models, simple LUR-like regression models were also developed to estimate yearly average exposure in transport (chapter 3.3). Mobile monitoring data was available from a personal monitoring campaign in 62 individuals, simultaneously measuring BC and GPS positions. Exposure in motorized modes can be predicted by using information on timing of the trip (peak, off-peak, weekend), degree of urbanization (highway, urban, suburban, rural), and instantaneous traffic intensity (vehicles per hour). For active modes timing and urbanization are significant predictors. Because FEATHERS combines all public transport modes, one constant concentration is applied here for all trips with public transport (3521 ng/m^3) although it is known that for example exposure in buses is much higher than exposure in trains (chapter 3.2).
Output from the activity-based model FEATHERS includes information on transport mode, timing, origin and destination. Individual trips are not assigned to a road network, so no information on the exact route is available. Because urbanization type is thus unknown, in chapter 3.3 we suggested using trip duration of motorized trips as a proxy for the use of roads with different degrees of urbanization (e.g. longer trips (>45min) are on highways for at least 45% of total time, whereas short trips (<30min) use highways only sporadically). For trips by bike or on foot, the urbanization type of the subzone of origin is extended to the whole trip. Datasets and model development are described in detail in chapter 3.3 and in appendix A.4.

4.3.2.4 Indoor air model

No parameterized indoor air model, like the INDAIR model or alternatives (Dimitroulopoulou et al., 2006; Gerharz et al., 2013) was applied because there is no detailed info on housing characteristics neither for the complete study area, nor for individual subzones. As outdoor particles are the largest contributor to indoor air pollution, an indoor/outdoor-ratio (I/O-ratio) was calculated and applied to estimate exposure in indoor microenvironments. The I/O-ratio is based on facade-measurements and simultaneous indoor measurements in 24 houses in the study area. Residences with tobacco smoke were excluded. Specific indoor sources of BC, like candles or cooking (LaRosa et al., 2002; Wallace, 2005), were ignored, although they were inherently included in the calculation of the I/O-ratio. An I/O-ratio of 0.76 was determined with indoor concentrations being lower than outdoor concentrations, and based on measurements in cold and warm seasons (appendix FIGURE A31). In warmer seasons, the ratio was higher (0.84) representative of better ventilation, although ambient concentrations were generally lower.

4.3.2.5 Integration of models: AB²C

Four data sources/models are used to predict personal exposure to BC: individual diaries from FEATHERS, hourly LUR models, the in-traffic exposure model, and an I/O-ratio (FIGURE 29). Predicted hourly BC concentration fields are combined with information on people's locations to calculate their minute-to-minute exposure (FIGURE 30). The activity-based model does not discriminate between indoor and outdoor activities; but as 87% of the time is spent in indoor environments (Klepeis et al., 2001), all activities (except trips) are assumed to be indoors and the I/O-ratio was applied to the ambient LUR predictions. When agents are traveling, the in-traffic exposure model representing yearly averages is applied taking into account transport mode, timing, location and duration of the trip. For touring activities, the in-traffic exposure model for active modes is used (these are activities where people are in transport but without a specific destination and with the same start and end point).

Several exposure metrics were calculated: for each modeled agent, (i) ambient concentration at the residence was calculated using the hourly LUR models for the relevant subzone, (ii) static exposure was calculated multiplying the ambient concentration with the I/O-ratio, and (iii) dynamic exposure was calculated making full use of the AB²C model, i.e. by including population mobility. The FEATHERS model simulates one diary for every agent in the population, for every day of the week, and the AB²C model can calculate exposures from these data. Alternatively, the FEATHERS model can produce a predetermined number of diaries for one individual or for a subset of individuals. The latter enables an estimation of a range of potential exposures for one agent using AB²C while meeting specific preconditions (subzone where the residence is located, household composition, household income, age of the oldest adult, age of the youngest child, the number of adults in the household, the number of cars in the household, age and gender, work status: homemaker or worker, possession of driver's license).

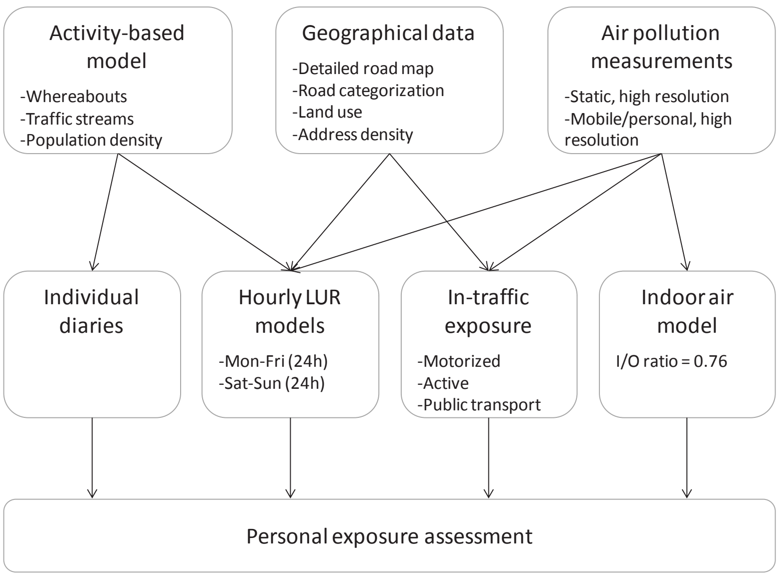


FIGURE 29: Integration of submodels to predict personal exposure to BC: the AB²C model.

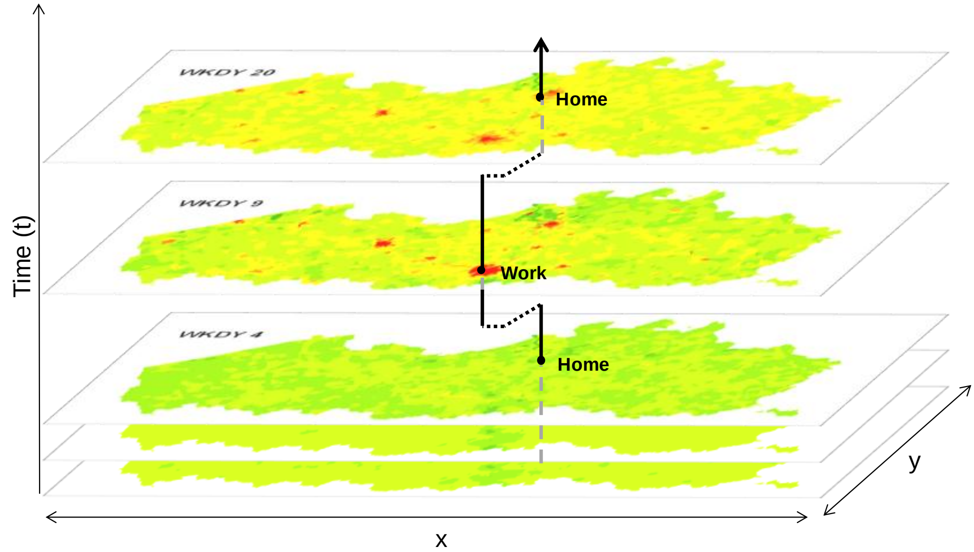


FIGURE 30: Example of personal exposure estimation for one agent using the AB²C model.

4.3.2.6 Validation dataset

A personal monitoring campaign was carried out in 62 volunteers (54 unique participants) from 31 families (27 unique families), exactly half of them were male (chapter 3.2). All were living in Flanders, the northern part of Belgium, in urban as well as in suburban or rural zones (appendix FIGURE A32). There is a slight overrepresentation of people working in a rural environment because 12 colleagues from our institute were recruited as well. All volunteers were between 20 and 60 years of age, but 10 participants were not working at the time of the personal monitoring campaign. Participants were asked to carry a portable aethalometer to measure BC (model AE51 (AethLabs, 2011)), an electronic diary and a GPS logger for 7 consecutive days to capture weekday-weekend differences in time-location-activity patterns. BC measurements had a time-resolution of 5-minutes, activities also had to be reported with an accuracy of 5-minutes, and GPS position was logged during trips on a 1-second basis. Measurements were done in May - July 2010 and December 2010 - February 2011. Up to three couples participated in the same week, and 4 couples

participated twice: once in summer and once in winter. As measurements were not always simultaneous, personal measurements were rescaled to account for changing background concentrations (chapter 3.2).
The characteristics of each real-life volunteer (N=54) were loaded in FEATHERS and 100 diaries were stochastically simulated for each weekday and for each model-agent, resulting in a synthetic population (54*700 modeled diaries in total). Characteristics that could technically be taken into account were limited (see 'preconditions' described in previous paragraph); and thus do not include subzone of work location, or time in transport. Again, from these diaries personal exposure to BC was estimated for each 24h-period using the AB²C model. The model provides a range of potential exposures for that individual, and also the likelihood of exposures above a particular level.
There is a small bias in our validation sample: volunteers in the monitoring campaign worked more (17% versus 13%), were less at home (65% versus 76%) and spent more time in transport (6.3% versus 4.8%) than the model-agents (appendix FIGURE A33). The modal split is rather similar between the modeled and the revealed diaries for car (57% versus 51%), active modes (31% versus 39%) and public transport (12% versus 10%).

4.3.3 RESULTS

4.3.3.1 Predicted and observed exposures: aggregated analysis

Average modeled 24h exposures compare well with measured values (FIGURE 31). But neither of the three exposure metrics can properly capture the full exposure range as experienced by volunteers (FIGURE 31). Ambient concentrations at home are more similar to measured personal exposures than static indoor concentrations. This is surprising since static indoor concentrations represent more the actual exposure, taking into account the majority of time is spent indoor. It is hypothesized that due to the high BC concentrations encountered in traffic, the ambient model overestimates exposure at home but this is counterbalanced by not taking into account increased exposure in traffic. Ambient exposure modeling overestimates exposure of people spending a lot of time indoors. Static exposure is in most cases lower than dynamic exposure, except for agents living in high concentration zones. Moving out of a highly polluted area may lower exposure; even if a trip is necessary (Dhondt et al., 2012b).

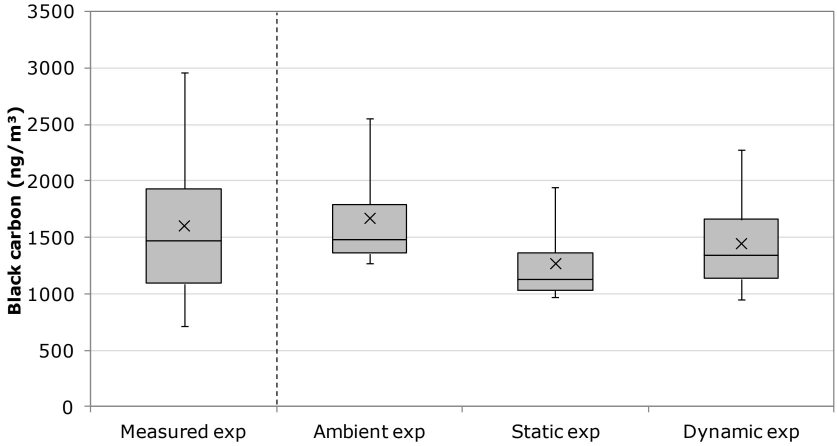


FIGURE 31: Daily average personal BC exposures measured in 62 volunteers, ambient exposure (modeled outdoor at home), static exposure (modeled indoor at home) and dynamic exposure estimates from the AB²C modeling framework. The box-and-whiskers represent 5th, 25th, median, 75th and 95th percentile; the X denotes the average.

An analysis on the level of single activity types reveals that the AB²C model overestimates exposure in transport and during touring activities. This also reflects the use of different transport modes in the model and the real-life sample, and a difference in location and timing of the trips. Exposure during social activities and shopping is underestimated by almost 50% because indoor-sources are not included in our model. Exposure at work is overestimated by 200 ng/m³, but this can be explained by the bias in our validation sample: too many participants work in remote areas. It holds for all activities and modes that the modeled and measured values are not entirely comparable as timing and location of the activities/trips might differ (appendix TABLE A15 and A16).

Pearson correlation between average modeled exposure and observed exposure in 62 volunteers is 0.452 using the dynamic exposure estimation; the root mean square error (RMSE) is 438 ng/m³. The correlation between observed exposures and static and ambient exposure is 0.451, with RMSE's of 537 ng/m³ and 489 ng/m³. Using the dynamic exposure estimation marginally lowers the uncertainty on the estimation, but the dynamic AB²C model is better in explaining the variability in the observations than ambient or static exposure models.

FIGURE 32 estimates the fraction of persondays above a certain BC concentration using both the AB²C model and personal monitoring. The fraction of people exposed to levels higher than 2250 ng/m³ (comparable to 1500 ng/m³ elemental carbon, conversion ratio estimated for the study area (Van Poppel et al., 2012a)) is zero when ignoring population mobility and assuming everyone indoors, whereas measurements show that 18% of the persondays have exposure levels above 2250 ng/m³. Dhondt et al. (2012b) did a similar estimation for Flanders using a dynamic population exposure model, resulting in 14.2% of the population being exposed to elemental carbon levels above 1500 ng/m³.

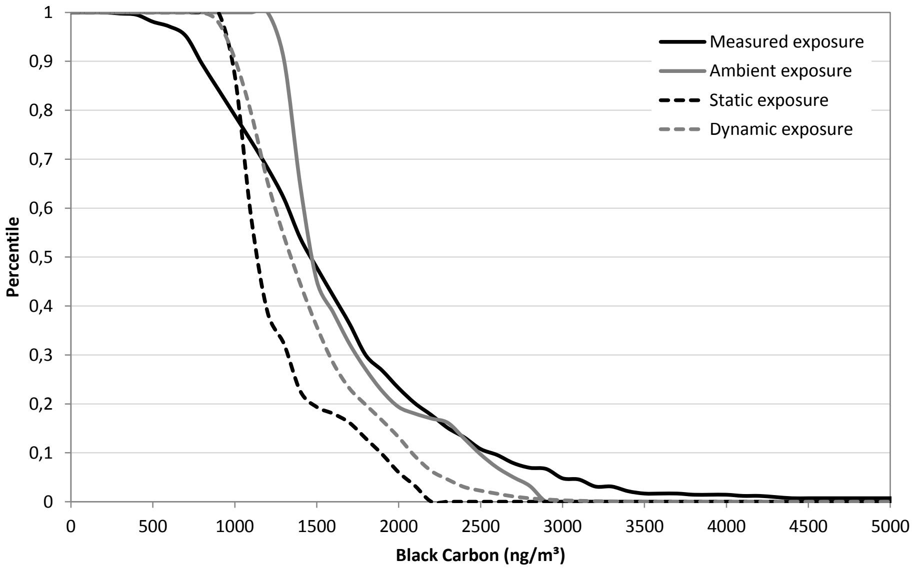


FIGURE 32: For every agent (real-life and modeled) annual average daily exposures are measured or estimated. The likelihood of 24h-averaged BC exposures above a particular level is presented for different exposure metrics (ambient, static and dynamic exposure estimates, and measured exposure).

4.3.3.2 Predicted and observed exposures: individual agents

Instead of focusing on the performance of the model on an aggregated level, namely for all 62 agents together, one can also analyze model performance for individual agents. The AB²C-model was run 700 times (100 times for each day of the week) to obtain a distribution of potential exposure levels for each of 62 synthetic agents with the same attributes (age, gender, work status, etc.) as the 62 participants whose personal exposure was measured (FIGURE 33).
Average modeled concentrations range between 1203 ng/m³ (household 15, person 2) and 2129 ng/m³ (household 2, person 2) using the dynamic exposure from the AB²C model. The interquartile range (IQR) is rather small with an average IQR of 352 ng/m³. Volunteers living in urban areas have the highest predicted average exposures to BC (FIGURE 33), and the largest IQR. The range between the minimum and maximum predicted exposure is much larger: 2306 ng/m³. The upper tails are caused by hypermobile agents; this includes agents performing recreational touring activities. From FIGURE 33 it is clear that the intra-individual variability in potential exposure levels exceeds inter-individual variability.

Eight volunteers participated twice in the personal monitoring campaign, and their measured personal exposure during both weeks differs up to 40%, even after rescaling to account for changing background concentrations. This reflects a different activity pattern, or indicates that volunteers came into contact with different sources. FIGURE 33 presents a distribution of daily predicted dynamic exposures, consisting of 100 Mondays, 100 Tuesdays, etc., whereas the measured exposure is averaged over one week to prevent single activities distorting average concentrations. In all cases, except for one volunteer, the measured exposure (temporal adjustments were made on measured values to represent annual average exposure) falls within the modeled exposure range. This one person has low average concentrations at home, at work and in transport which the model cannot capture; the I/O-ratio of 0.76 is an overestimation for this one volunteer at home (I/O-ratio of 0.59 measured for this specific house).

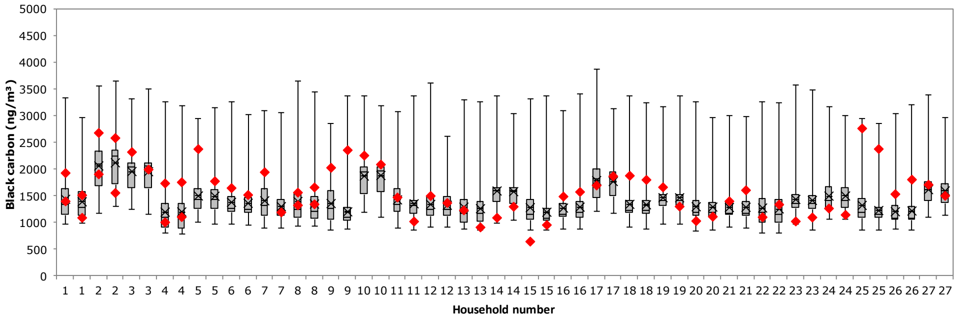


FIGURE 33: Predicted dynamic exposure using AB²C for every volunteer; the diamond shows the observed exposure (adjusted for changes in background concentrations). All observed exposures are within the predicted ranges except for household 15, person 1. Households 2, 3, 10, 17 and 27 live in an urban area. The box-and-whiskers represent minimum, 25th percentile, median, 75th percentile and maximum; the X denotes the average.

4.3.4 DISCUSSION

In the AB²C modeling framework a bottom-up approach was pursued, starting from the exposure of individual agents in specific microenvironments and aiming to evaluate 24h-exposure of a full population. Persondays with high levels of predicted exposure were traced as the upper tails are the most important portion for many risk assessments. This is also the first paper to actually validate the final outcome of a personal exposure model using novel monitoring techniques.

4.3.4.1 AB²C model

The AB²C model consists of several submodels, a.o. an activity-based model, hourly LUR models, an in-traffic exposure model and an indoor air model. Single submodels can be replaced with other models, making this a flexible framework. The hourly LUR-models could be replaced with emission and dispersion models (Beckx et al., 2009b), other agent-based models or revealed GPS tracks could be used instead of whereabouts from FEATHERS (de Nazelle et al., 2013;

Gerharz et al., 2013; Newth and Gunasekera, 2012). Separate modules can be adapted to use AB²C for additional air pollutants.
Seasonal differences are not taken into account in neither of the submodels; although personal BC measurements showed that the exposure of one individual can vary significantly between weeks or seasons. Time-activity patterns also differ between seasons (Isaacs et al., 2013; McCurdy and Graham, 2003). But because the FEATHERS model cannot distinguish between different seasons, our modeling framework predicts annual average exposures. The hourly LUR models, the in-traffic exposure model, and the indoor air model are tailored to the needs of the activity-based model, and thus none of these models are currently set up to be season-specific.

Activity-based models were previously used in population exposure studies: Beckx et al. (2009b) used ALBATROSS in the Netherlands, Hatzopoulou et al. (2011) used TASHA for the Greater Toronto Area, Lefebvre et al. (2013a) and Dhondt et al. (2012b) used the same activity-based model as in this study, FEATHERS, but they focused on population exposure. Activity-based or agent-based models are still rather scarce, but promising environmental and health applications using such type of models are emerging (Int Panis, 2010).

4.3.4.2 Modeling exposure on fixed locations

The FEATHERS model builds a diary and assigns activities to a subzone. The exact address or position of the modeled activity is therefore not specified and exposure on fixed locations cannot be directly determined using high resolution LUR data. Therefore exposure was calculated for a random selection of addresses in each subzone; the median concentration was then assumed to be representative for exposure on all addresses in the subzone. The true value can deviate from this median for a specific house, e.g. a building next to a major road. For the validation dataset, it would have been possible to use the precise geographic location of the home, but then this would not be a validation of the complete model chain because exact residential locations are not known from FEATHERS.

4.3.4.3 Modeling in-traffic exposure

In other exposure studies, in-traffic personal exposures are either ignored (Hatzopoulou and Miller, 2010), concentrations were modeled (Dhondt et al., 2012b) or measured (Beckx et al., 2009b) at fixed stations near busy roads, or dispersion or LUR models were used to estimate exposure while traveling (Marshall et al., 2006; Mölter et al., 2012; Setton et al., 2011). For the AB²C-model, dedicated in-traffic exposure models representing annual average concentrations were developed for the study area and incorporated in the model (chapter 3.3). It was shown before that the performance of the in-traffic model for motorized modes could be further improved when traffic intensity is also taken into account. To be able to apply this extended version of the in-traffic exposure model in AB²C, traffic intensities would have to be calculated by assigning all the single trips on a road network. For the moment, the information on which agent makes which trip is lost when all trips predicted by FEATHERS are assigned to a road network, keeping only aggregated trip information. Assigning a single trip to a road network is also possible, but then traffic flow dynamics cannot be included. From chapter 3.2, it is known that exposure in traffic is responsible for 21% of total exposure to BC. As the difference between the average predicted exposure and the observed exposure is often larger than 21%, the application of a better in-traffic exposure model will not eliminate all deviations. It is also shown that with the current in-traffic exposure model spending more time in transport, leads to higher total exposures. However, spending a lot of time in transport is not a necessary condition to be highly exposed: personal exposure can be elevated without being in transport for a long time, i.e. when living in a polluted subzone.

4.3.4.4 Modeling indoor concentrations

In 24 residences of participants in the personal monitoring campaign, indoor and outdoor measurements were performed that resulted in an I/O-ratio of 0.76. This ratio is in line with values reported in previous studies and in other regions (LaRosa et al., 2002; Lunden et al., 2008; Stranger et al., 2008; Viana et al., 2011; Wichmann et al., 2010). Lunden et al. (2008) compared several studies and concluded that I/O-ratios for BC were primarily at or <1.0. More recent studies confirm these numbers (Viana et al., 2011; Wichmann et al., 2010); Wichmann et al. (2010) even found exactly the same ratio of 0.76 for soot. The use of an I/O-ratio instead of a parameterized indoor air model is a strong simplification and will result in over- and underestimations of exposure in certain microenvironments, e.g. restaurants, pubs, environments with tobacco smoke, mechanically ventilated buildings with air filters, etc. (Brown et al., 2012; Wichmann et al., 2010). Comparable to the inclusion of the precise geographical location of homes in the AB²C model validation, I/O-ratio's specific for each home could have been used. Again, it was preferred not to do this, to be sure to validate the complete modeling framework.

4.3.4.5 Model validation

The validation dataset comprises revealed diaries, exposures and GPS routes for 62 volunteers, and suffers from biases in the participant selection. Participants were urged to follow their habitual daily activities, but still many volunteers did not experience an annual average week: activities and locations visited were specific for that week, they came into contact with specific sources of BC, or they spent more time in transport than they usually do. To avoid this, longer personal monitoring campaigns would be necessary. Nevertheless, despite these disadvantages, the validation dataset used in this study is relatively large, both in number (54 unique participants) and in duration (7 consecutive days per participant) (Duan, 1991; Gerharz et al., 2013; Mölter et al., 2012).
For 54 unique individuals, different exposure metrics were computed based on a very limited number of characteristics of an individual (i.e. those used by the FEATHERS decision trees). Mean exposures were not that different using the different exposure metrics, but the exposure contrast was larger using the dynamic method. Dynamic exposure takes into account trips and activities performed at locations other than the home, and this exposure metric predicted observed personal exposure slightly better than ambient or static exposure measures. However, the added value of a dynamic model lies in the possibility of

detecting short term peak exposures, high exposure activities and exposure contrasts rather than in only reproducing average exposures. It should be noted that what is called the ambient/static approach in this study is based on one concentration per subzone, which is less detailed than in recent epidemiological studies that are based upon address level.

For all participants in the personal monitoring campaign, 700 diaries (100 diaries for every day of the week) were produced by the activity-based model and then, using the AB²C model, this resulted in distributions for BC exposure for every agent. This large intra-individual variability in time-activity patterns is largely ignored in existing exposure models. Given our results, we think it should be an important aspect of these models, something that was also stressed by Isaacs et al. (2013). The large whiskers in Figure 5 represent the exposure of a minority of hypermobile agents (people spending a lot of time in transport): this is not an artifact of the model, but this is indeed observed in field studies as well (Klepeis et al., 2001).

The AB²C model showed relatively good model performance, but in some cases there were larger differences between the predicted and observed concentrations. An explanation is that the LUR models and the in-traffic exposure model are based on the normal distribution and therefore will perform best when predicting concentrations close to the mean (similar to (Mölter et al., 2012)). Extreme conditions could be predicted better by using a stochastic I/O-ratio or by including stochastic parameters in the in-traffic exposure model. This would not influence average personal exposures based on hundreds of diaries, but the variation would be larger.

4.3.4.6 Limitations

The rather coarse resolution, 2386 subzones of 5.7 km² on average, of the FEATHERS predictions contrasts with the highly detailed air pollution surfaces produced by the LUR models. The activity-based model FEATHERS could be applied to zones smaller than the subzone-level (the so-called ‘building block’ level), but the essential underlying data is not available on this level (e.g. number of work places, composition of the population). By predicting concentrations for existing addresses in a subzone, the exposure of individuals in a subzone was approximated.

The fact that the validation dataset is not completely independent of the calibration dataset is a drawback of the study. The in-traffic exposure model is built with travel information from the same 62 volunteers that serve as a validation, although the in-traffic models are estimated with information of all individuals and is then applied to single trips. In the development of the hourly LUR models, measurements at 63 sites were used to calibrate the model. From these 63 sites, 21 sites were home locations from participants in the personal monitoring campaign; all the other measurements came from an independent purpose-designed monitoring campaign. The I/O-ratio applied to all activities excluding traveling, is estimated with information from 24 residences: these are all home locations from the volunteers serving as validation. The I/O-ratio of 0.76 showed quite some differences between homes and between days; using the average ratio in the AB²C model will not bias the validation for single agents. In the future, the AB²C model or single submodels should be validated with a fully external validation dataset.

4.3.4.7 Future prospects

In the future traffic should be assigned more accurately, e.g. using micro-simulation models, to make FEATHERS variables more predictive in LUR models, but applying micro-simulation models on a national scale is not yet possible. The inclusion of FEATHERS variables in the hourly LUR models would open up a myriad of possibilities in calculating the effect of policy scenarios that, by changing the time-activity pattern of individuals, affect concentrations, personal exposure and health.

Steinle et al. (2013) designed a conceptual model to use individual exposure estimates to derive population-wide exposure estimates and investigate their implications for public health. The AB²C model has this potential to calculate exposure for a complete population or for a subset of the population using only very limited input-data. Not only long term exposures can be calculated or the fraction of a population exposed to levels above a certain threshold, but also the frequency of short term exposures above a certain level can be determined. The latter may bring new opportunities to epidemiologists: studying the effect of frequently repeated but short exposure peaks on long term exposure and health.

5. DISCUSSION AND FURTHER RESEARCH

5.1 DISCUSSION

5.1.1 MEASURING PERSONAL EXPOSURE

5.1.1.1 Air quality sensing

Air quality measurements with portable fast response instrumentation are essential to understand personal exposure to air pollution. Unfortunately, devices or sensors that are cheap, are not overburdening participants, and reliably measure a selected air pollutant, are not yet developed. Micro-aethalometers are used in a personal monitoring campaign in this PhD: this device measures BC, is small and mobile, but rather expensive. Micro-aethalometers can be transported in a backpack or handbag, as long as the inlet of the monitor is exposed to ambient air. The inlet should be placed as close to the breathing zone of the individual as possible to limit possible confounding (Adams et al., 2009; Fruin et al., 2004). To extend battery lifetime, it was necessary to measure on a lower time-resolution (5-min), combined with an intermediate sampling rate to limit audible noise. Application of a smoothing algorithm is not required when measuring on this time resolution: only a small percentage (approximately 2%) of the readings were negative (Hagler et al., 2011). Filters had to be replaced twice per week to prevent saturation and to maintain data integrity. Still, additional post-processing on the data was necessary: 1) all data showing an error code were excluded, except for low battery events; 2) BC readings were excluded when the attenuation was above 75; and 3) corrections for device specific deviations were made based on an intercomparison of all devices. Overall, the use of a mobile platform for weeklong personal exposure assessment in 62 volunteers was innovative both in setup and in size (Steinle et al., 2013).

In complement to the personal measurements, fixed outdoor measurements were done at the residence of the couples. It allowed to calculate I/O-ratios whenever participants were at home with their personal aethalometer.

5.1.1.2 Time-activity patterns

Time-activity diaries used to be collected in paper format, but electronic diaries are slowly gaining in popularity. Small electronic devices, PDA's or smartphones, make data recording more flexible in space and time, and minimize the burden for study participants (Delfino et al., 2010). In this PhD, a PDA application called PARROTS was used for the registration of weeklong time-activity diaries in 62 participants (Kochan et al., 2010). An electronic diary enforces all attributes of executed activities to be provided on a 5-min resolution and it includes consistency checks; accordingly it results in high-quality data with a non-response of zero. Fewer drop-outs were registered in case of the PDA survey, indicating that the burden for filling in this kind of survey is lower in comparison with the traditional paper-and-pencil approach (Kochan et al., 2010). The reported number of executed trips is more stable throughout a survey using PARROTS and on average more trips per person were reported for surveys using PARROTS compared to the paper-and-pencil method (Kochan et al., 2010).

Movement of participants can be tracked with a GPS, and the space-time path or time-activity pattern can be reconstructed afterwards (de Nazelle et al., 2013; Gerharz et al., 2009). Problems include interruption or inaccuracy of the GPS signal due to the number and constellation of satellites and the built environment, or inaccuracy of the maps that the GPS coordinates are linked to (Marchal et al., 2005). In studies measuring GPS only, single trajectories need to be analyzed and annotated using semantic trajectories data mining techniques (Bohte and Maat, 2009; Wu et al., 2011a), while in this PhD the transport mode and trip motive were reported by the volunteers. Nevertheless, GPS tracking, both with and without activity annotation, has the potential to significantly reduce respondent burden. In the current study, the GPS signal has a time resolution of 1 second, leading to a temporal resolution mismatch with the 5-minute aethalometer measurements and reported diaries.

5.1.1.3 Personal monitoring campaign

Initially, 8 couples were recruited to measure personal exposure to BC for 7 consecutive days; one couple was monitored per week. These couples were characterized by a difference in activity pattern between both partners: a full-time worker and a homemaker. Differences in weeklong BC exposure between households (because of changing background concentrations) have been found to be larger than between partners of one household (although these differences also amount to a maximum of 30%), highlighting the challenges of up-scaling from individual to population exposure. In this effort, 38 extra participants (19 couples) were enrolled and 4 couples participated a second time, but this time in a contrasting season.

Thirteen different measurement devices were used; before and after each measurement campaign an intercomparison of the devices was made (appendix FIGURE A1). Devices compared very well, with $R^2 > 0.9$. The slope of the regression function was used to correct personal and fixed measurements during the field campaign (corrections were between 1% and 23%).
As not all couples measured BC in the same week, adjusting for non-simultaneous measurements is necessary if studying the impact of e.g. transport mode on exposure. Whether the variations in background concentrations are additive, multiplicative, or a combination of both methods is an unsolved question, and probably depends on the pollutant and the location. In this PhD, we compared different methods and proposed a combination of the additive and multiplicative method (appendix A.1). The main conclusion was that adjustments, independent of the rescaling method, were always smaller than differences between different activities and between different microenvironments.

Traveling contributed most to variability in personal exposure to BC between people exposed to the same concentrations at home: Volunteers spend on average 6% of their time in transport, but this accounts for 21% of their personal exposure, and to 30% of the inhaled dose. Unfortunately travel time proved to be an unsatisfactory parameter to estimate personal exposure to BC

pollution because of the many factors influencing exposure in traffic (transport mode, timing and location of the trip, traffic intensity on the road). Finding a relationship between travel time and BC exposure would have been very interesting as travel time is available from many exposure models, and it is easy to query in larger cohorts.
For now, inhaled doses are calculated using minute volume assumptions linked to different activity levels and activity types, but this could be improved by directly measuring a human health parameter for level of effort.

The combination of portable air quality measurements at a high temporal resolution, and an electronic diary with GPS enabled to identify activities, microenvironments or locations contributing disproportionally to the exposure. Few studies so far really covered the heterogeneity of a person's everyday microenvironments; most studies monitor air quality in a few selected microenvironments only (Brown et al., 2012; Steinle et al., 2013). Since we demonstrated that the exposure of two individuals living at the same location can differ by as much as 30% (as a maximum), using modeled or measured concentrations at the place of residence alone is neither accurate nor sufficient.

5.1.2 MODELING PERSONAL EXPOSURE

5.1.2.1 Activity-based model

Activity-based models have been used in the past for population exposure assessment. Shiftan (2000) was the first to describe the advantages of activity-based modeling for air quality purposes over traditional four-step models. Beckx et al. (2009c) and Hatzopoulou et al. (2011) applied local activity-based models to estimate traffic emissions and population exposure to several air pollutants in the Netherlands and Toronto. The activity-based model for Flanders FEATHERS was first used by Dhondt et al. (2012a) to calculate population exposure and health impact, but without proper validation and not focusing on personal exposure. Activity-based models originate from traffic science which is an advantage as these models focus on traffic.

The activity-based model for Flanders, FEATHERS, is not specifically built for air pollution exposure assessment. This shows in the choice of microenvironments, e.g. no distinction between indoor and outdoor activities, sleeping is not predicted as a distinct activity, trips by train, bus or metro are grouped under one category.

The activity-based model can be improved in multiple ways to simulate diaries more realistically, and by extension improve the personal exposure estimation. In the current model, a single trip cannot make use of multiple transport modes; instead the main transport mode of a trip is selected. Problematic for weeklong exposure is that there is no association between the same person or agent on different days of the week (Monday-Sunday). Person x on different days is as much alike as a new agent with the same input-characteristics.

The concept of subzones or TAZs (2386 in the study area) used within the FEATHERS transportation model eliminates intrazonal traffic. Trips that originate and arrive in the same subzone are predicted in individual diaries, but these trips cannot be assigned to a road network in the traffic assignment step. As a result local traffic is not present in the traffic streams used as input in the hourly LUR models. On the other hand, remaining traffic is assigned to a dense road network using an equilibrium assignment, this means that congestion effects and rerouting are taken into account, which is an improvement over previous studies (Beckx et al., 2009c).

5.1.2.2 Hourly LUR models

FEATHERS variables are occasionally included in the hourly LUR models, but it was anticipated that especially traffic intensities would emerge more frequently in the air quality models. BC is a traffic-related pollutant, with steep decay rates when moving away from roads: local traffic intensities thus impact local ambient concentrations (Karner et al., 2010; Zhu et al., 2002). The traffic assignment used in this PhD is apparently not sufficiently accurate: we are using a macroscopic traffic model, but we actually need a microsimulation traffic model.

Unfortunately, microscopic models cannot yet be applied to our complete study area because of computational and data constraints.

High variance inflation factors (specified as VIF > 3 (Beelen et al., 2013; Eeftens et al., 2012); or as VIF > 5 (Johnson et al., 2010)) are a problem since they indicate multicollinearity in regression models. In the hourly LUR models elevated levels for the Cook's D were found but they were never larger than 5.
An important advantage of LUR models is the good cost-benefit ratio compared to dispersion models; but the need to build 48 separate LUR models might weaken this point. In our experience, hourly LUR models are not more data-demanding than annual LUR models, except for the models with dynamic covariates but those were not retained in the AB²C model. Time-resolved measurements should be available, but the use of an active monitor does not necessarily require more supervision than passive samplers once in place. Estimating 48 LUR models obviously takes more time than developing one annual LUR model, but model development can be largely automated and running the script takes only seconds, even on a laptop computer. Dispersion models have the disadvantage of needing certain input variables, e.g. meteorological variables. Land use regression models are easier to use and quicker to implement as they can be built with available data.
Dispersion models typically use raster grids (with one grid cell being 1km² to several km²): dispersion models are then unable to accurately predict within-city variability in air pollution, something that is possible with LUR models. Recent developments in dispersion modeling increased the spatial resolution of these models (adding an irregular line source following grid using IFDM; inclusion of a street canyon module). One example is a dispersion model for NO_2 that was developed for Flanders, and validated with passive NO_2 measurements (measurements from campaign (i) in chapter 4.1) (Lefebvre et al., 2013b). This latter paper shows that the extended dispersion model is indeed much better in estimating within-city variability in concentrations compared to a gridded dispersion model.

5.1.2.3 Indoor air model

People are often in indoor environments. It is estimated that a typical person is indoors approximately 90% of the time (Klepeis, 2006). The activity groups defined in the FEATHERS model do not explicitly differentiate between indoor and outdoor microenvironments; therefore all activities are hypothesized to be indoors (except for traveling).

Complex indoor air models, for example the mass-balance model approach, require much data: ventilation properties, air exchange rates, filters used, indoor sources, room volume, deposition rates, etc. (Gerharz et al., 2013; Lunden et al., 2008). Because indoor concentrations depend on many factors and conditions, they vary considerably between houses, which also appeared from the measurements reported in this dissertation. It is impossible to collect these data on a population level, but assumptions could be made based on a small sample where extensive measurements took place or based on values reported in peer reviewed literature (Gerharz et al., 2013).

5.1.2.4 Validation of AB²C

Different models are integrated in one overall framework: the AB²C model. This model predicts long term exposure to BC for the Flemish population by taking into account population mobility and time-activity patterns. This improves on current methods that mostly ignore the fact that people spend time away from home and are exposed to concentrations that differ from ambient concentrations at a fixed location. Using our approach, peak exposures, or highly exposed individuals or groups of individuals can be identified; and the origin of the elevated exposure can be traced. Also, because of the stochastic nature of the FEATHERS model, intra-individual variability in activity pattern and in exposure can be modeled.

Compared to previous attempts to model exposure to air pollution using activity-based models (Beckx et al., 2009a; Dhondt et al., 2012b), the AB²C model focusses on personal exposure rather than on population exposure. These population exposure models estimate exposure using ‘personhours’ spent in each zone, and it is not possible from this output to calculate the accumulated exposure of an individual. The added value of a model focusing on personal exposure are the opportunities it creates for individualized mitigation policies, the calculation of health impact and linked costs, and the identification of people exposed to levels exceeding a certain threshold.

The newly developed AB²C framework can also be compared to a more traditional way of calculating population exposure: by using dispersion models and static address information. For Flanders, daily EC concentrations were determined by using a MIMOSA-AURORA-IFDM model chain (emission and dispersion modeling) for the year 2007 (Lefebvre et al., 2011). This concentration map was combined with static address data for the complete population resulting in a population exposure distribution (FIGURE 34). This exposure distribution is confronted with the measured personal exposures as reported in chapter 3 (adjusted for changing background concentrations and thus representative for annual average exposure); while bearing in mind that this sample was not completely representative for the Flemish population. For the majority of the people, exposure is underestimated, most likely because population mobility and exposure during traveling is not taken into account (FIGURE 34). Overall the curves are rather similar, indicating that on a population level the traditional way of calculating exposure is a good approximation for real exposure, despite the small underestimation.

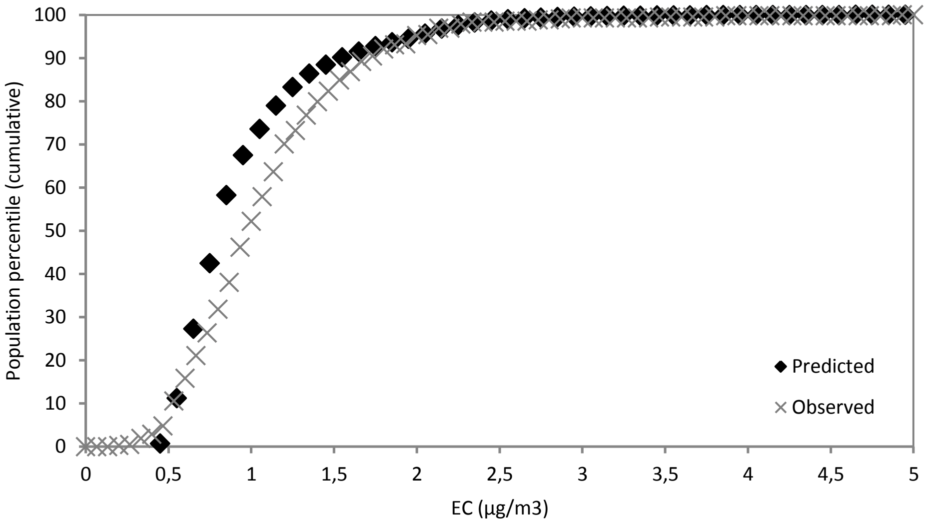


FIGURE 34: Comparison between predicted exposures, and observed personal exposures in 62 volunteers in Flanders (chapter 3). Personal measurements were done for BC; for the purpose of this figure, BC concentrations were converted to EC (BC = 1.5 * EC). In total, 418 daily personal exposures are reported.

On the disaggregated level of an individual, the performance of the AB²C model is compared to real-life personal measurements. For every volunteer that participated in the personal monitoring (chapter 3), personal and household characteristics (subzone where the residence is located, household composition, household income, age of the oldest adult, age of the youngest child, the number of adults in the household, the number of cars in the household, age and gender, work status: homemaker or worker, possession of driver's license) were collected and loaded into FEATHERS. Based on these characteristics, 100 virtual agents were created per day (Mon-Sun, 700 agents in total) and for every agent a diary was simulated. This was done for all 54 unique participants. Then personal exposure was calculated using the AB²C model, and estimated concentrations were compared to observed BC exposure. Significant intra-individual variability was observed resulting in wide exposure distributions. In chapter 4.3 this personal exposure distribution is compared to the weeklong average exposure of the real-life individual. For every individual a more detailed analysis is possible comparing observed daily concentrations to the exposure distributions. In FIGURE 35, the exposure distribution for one agent is presented, and is compared to measured exposures on different weekdays. As can be seen, both the modeled and the measured exposures show relatively much variation between different days. The figure represents an 'annual average day', so changes in background concentrations are not yet incorporated, but this would even increase the variability. Predicted exposure is lognormally distributed, with a tail to the right where health risks are most important (Isaacs et al., 2013).

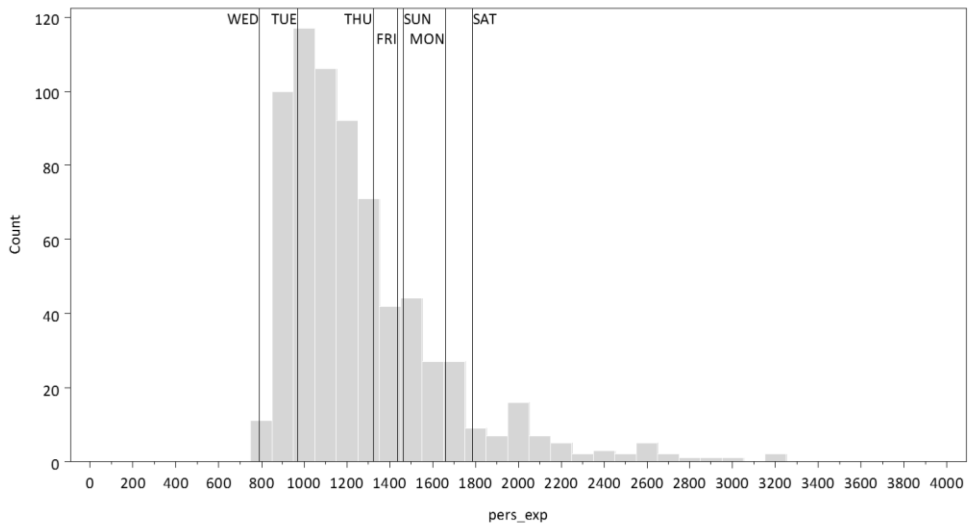


FIGURE 35: Personal exposure to BC (ng/m³): observed (vertical lines) versus modeled (histogram) using AB²C (Household 22, Person 2 on FIGURE 33)

5.1.2.5 Comparison of AB²C with existing exposure models

SHEDS, pCNEM, and the activity-based modeling frameworks discussed before (Beckx et al., 2009a; Dhondt et al., 2012b) are all examples of previously developed population exposure models. STEMS, BESSTE, MEEM and EMI are examples of models that estimate personal exposure (see chapter 2 for a detailed description of these models). All these models contribute to a more realistic estimation of exposure compared to the use of concentrations measured with fixed air quality monitors. Still many of these models make unrealistic assumptions, some of which we tried to tackle in this PhD.

- An approach that is often adopted in population exposure models is the use of activity databases. This is an improvement over the use of a non-mobile population, but in most cased the activity database is deterministic in nature: persons with the same characteristics will have the same agenda, largely ignoring both inter-individual and intra-individual variations. Activity databases often lack information on the geographical location of activities, and concentrations are assumed per microenvironment. This means that a residence in an urban area will have the same predicted concentration than a residence in a rural environment. By using individual diaries estimated by the activity-based model FEATHERS, AB²C improves on both of these points.
- Estimating exposure while traveling is more difficult than estimating exposure for fixed locations because the location and conditions are constantly changing. In some studies, exposure while traveling is completely ignored (e.g. in pCNEM, and by Hatzopoulou and Miller (2010)). Other approaches include the use of dispersion or land use regression models to estimate concentrations on roads. Though these models are not intended to estimate concentrations close to emission sources, partly also because these models are built using measurements on fixed locations away from traffic. This approach also ignores that in-cabin concentrations can differ from concentrations on a road. For AB²C, an in-traffic personal exposure model for Flanders representing annual averages was estimated, taking into account transport mode, timing, street and traffic factors.
- Indoor BC concentrations are generally lower than outdoor BC concentrations. Complex indoor air models are used in SHEDS and EMI, in MEEM and in the personal exposure model for Münster. Indoor air quality models either need specific data for each individual building not available on an aggregated level, or values for each parameter need to be retrieved from a limited set of buildings and then mean values are used in the model. The second method is an approximation and errors can be large for individual houses. The AB²C model uses an I/O-ratio that is applied to all activities

(except traveling). The I/O-ratio is based on measurements in 24 houses in Flanders, but we did observe differences between houses.

- Neither of the aforementioned models is properly validated using exposure measurements; the validation was often limited to the evaluation of submodels. Exposure models that estimate personal exposure for every individual in a population, need to validate time-location-activity diaries as well, next to the validation of the air quality model. Most current personal exposure models (a.o. EMI, MEEM, Personal exposure model Münster) are based on revealed diaries (using paper diaries or GPS) and cannot be generalized to a complete population. By using an activity-based model, diaries are generated stochastically for a synthetic population, and thus additional collection of diaries is not necessary: using AB²C personal exposure can be estimated on a population scale. Moreover, the AB²C model is validated through personal monitoring.

5.1.3 WEAKNESSES

Personal monitoring was done in 54 unique individuals, of which 8 participated in two seasons. In total, a dataset of 62 weeks was available consisting of BC measurements, a time-activity diary, and GPS tracks. This sample is large compared to efforts from other authors (Broich et al., 2012; Delgado-Saborit, 2012; Steinle et al., 2013), but is still limited as this sample cannot be regarded as representative for a complete population. There is a bias in our recruited sample because too many volunteers worked in a rural area (nine colleagues from our own institution), most had an office job, and all workers worked indoors. In line with many exposure and health studies, our participants were biased toward higher education. Most volunteers participated in a cold season, only 16 persons participated in a warm season. BC measurements in different seasons and in different weeks are adjusted for changing background concentrations, but changes in the time-activity pattern cannot be accounted for (e.g. more outdoor activities in the summer).

Over the last years many novel sensors have become available measuring movement (accelerometry, GPS) or body functions (heart rate, ventilation, galvanic skin response, etc.). In this PhD, no proxy for physical activity was measured, making it impossible to truthfully report inhaled doses. Assumptions on minute volume were made in chapter 3.2 based on activity type and gender (Allan and Richardson, 1998). Inhaled doses can be linked directly to health endpoints making this a valuable parameter to measure in future personal monitoring studies.
It really is a shame that we did not measure any health parameter in the participants of the personal monitoring campaign. Several non-complex and non-invasive biomarker measurements associated with acute exposure to BC would have been possible, e.g. exhaled NO, retinal pictures, blood pressure (Delfino et al., 2006; Lin et al., 2011). These biomarker measurements could have demonstrated that health studies really benefit from detailed exposure assessment.

The coarse resolution of the FEATHERS predictions contrasts with the highly detailed air pollution surfaces produced by the LUR models. In the FEATHERS platform, 2386 TAZs or subzones were defined in Flanders (including Brussels) with an average surface area of 5.7 km². The activity-based model FEATHERS could be applied to zones smaller than the subzone-level ('building block' level), but the essential underlying data is not available on this level (e.g. number of work places, composition of the population). By predicting concentrations for existing addresses in a subzone, the exposure of individuals in a subzone was approximated.

Error propagation is an important issue in the use of linked models (Gulliver and Briggs, 2005). More sophisticated exposure models like AB²C do not necessarily improve exposure estimation. Validation across a wide range of conditions, across different time periods and in multiple individuals is therefore required.

The validation dataset used in this PhD is not completely independent of the calibration dataset used to develop some of the submodels. The in-traffic personal exposure model is built with travel information from the same 62 volunteers that serve as a validation, although the in-traffic models are estimated with information of all individuals and is then applied to single trips of virtual agents. In the development of the hourly LUR models, measurements at 63 sites were used to calibrate the models. From these 63 sites, 21 sites were home locations from participants in the personal monitoring campaign; all the other measurements came from an independent purpose-designed monitoring campaign. The I/O-ratio applied to all activities excluding traveling, is estimated with information from 24 residences: these are all home locations from the volunteers serving as validation. The I/O-ratio of 0.76 showed quite some differences between homes and between days; using the average ratio in the AB^2C model will not bias the validation for single agents. In the future, the AB^2C model or single submodels should be validated with a fully external validation dataset.

5.2 FURTHER RESEARCH

5.2.1 METHODOLOGICAL CHALLANGES

To refine exposure estimation, the FEATHERS activity-based model should be able to predict diaries on a much finer scale than the subzone-level. Using this higher spatial resolution, traffic assignment will improve, and traffic streams will be more realistic. It is expected that traffic intensity will then be included in the hourly LUR models for BC. Currently, the spatial resolution or granularity of FEATHERS is already improved compared to previous releases (Dhondt et al., 2012a; Lefebvre et al., 2013a): the number of zones has increased from 1145 to 2386, and the average surface area of a zone has halved. Bottlenecks for further refinements are data availability and privacy issues.

The absence of FEATHERS variables in many hourly LUR models, currently limits the possibilities of using the AB²C framework for scenario assessment. Scenarios that impact the activity pattern of individual agents, will not affect the air quality; e.g. when calculating the effect of more teleworking, this will not impact BC concentrations through reduced traffic intensities. All scenarios that change the activity pattern of individuals will nevertheless change the exposure of people due to changes in the time-space path, and independent of changes in the air quality.

When developing the hourly LUR models, multiple issues came up. The rather short sampling period might be responsible for some erroneous or not generalizable measurements at certain locations for some hours. This probably limits the validity of the models presented in chapter 4.2. It is advisable to repeat measurements in multiple seasons, and to measure for longer continuous periods on each location, especially in the case that hourly LUR models are built. However, measuring BC using micro-aethalometers requires frequent filter changes, and the equipment is expensive, leading to a costly monitoring campaign. Cheaper, more reliable sensors may solve this constraint in the near future.

Models for succeeding hours showed overlapping significant covariates, pointing to the possibility of combining hours in groups. Grouping of hours could either be based on the correlation between measurements on different hours (but this does not guarantee similar air pollution sources), or alternatively it could be based on the similarity of the developed hourly LUR models. It should be investigated further how to combine models and whether it is useful to group them.

In this PhD, a personal exposure model is developed using several submodels, a.o. an indoor air model and an in-traffic exposure model. Single models can be substituted using alternative techniques, for example the in-traffic concentrations could be calculated by overlaying a LUR model with single trips; or indoor air quality could be assessed by using a mass-balance model approach. In the past, several researchers used an emission and a dispersion model instead of a LUR model to estimate concentrations on fixed locations (Beckx et al., 2009a; Dhondt et al., 2012b; Hatzopoulou et al., 2011).
The AB²C modeling framework can also be extended with additional submodels. An inhalation module to estimate dose or 'internal exposure' would definitely enrich the model outcome and further improve its relevance for environmental epidemiology. In chapter 3.2 it was shown that breathing rates can be very relevant: if traveling contributes approximately 21% to exposure, it accounts for 30% of inhaled dose. The AB²C model could be extended with a full grown inhalation module, coupling breathing rates to activities, transport modes and a person's characteristics. The extended AB²C model should then be validated in a personal monitoring campaign that also registers physical activity levels in volunteers (using heart rate monitors, face masks measuring ventilation, novel physiological sensors, or 3D-accelerometry).
The resulting exposure or dose estimates can be transformed to health effects in individuals by applying appropriate exposure-response or dose-response functions (Dhondt et al., 2013; Newth and Gunasekera, 2012). Health effects can then be converted to costs and serve as valuable information for policy makers.
Though, adding or replacing submodels should be done with caution. Complex models can become more of a black box, making it more difficult to draw unambiguous conclusions. Hence, complicating models does not necessarily enhance the validity of the model outcomes (Gulliver and Briggs, 2005).

The AB²C model is implemented and validated for BC. The added value of a personal exposure model that takes into account population mobility will be different for different pollutants. For pollutants that are relatively stable over space and time in Flanders, like PM_{10}, personal exposure will not drastically change if trips and out-of-home activities are not accounted for. To implement and validate the current AB²C model, it is necessary that the pollutant under investigation can be measured with a high temporal resolution, both in a fixed and on a mobile platform: devices that have this possibility are currently rare. Newly developed sensors could fill this gap and open up novel opportunities. Finally, it is not worth the effort to model exposure in great detail, if there is absolutely no toxicological evidence or even an indication that the pollutant is harmful to people. Pollutants that would make good candidates in future personal exposure models are NOx, SO_2, ultrafine particles, VOC, PM composites (EC, OC) and metals (e.g. Cu, Fe and Zn (non-tailpipe emissions from road traffic)).

5.2.2 OPPORTUNITIES FOR EPIDEMIOLOGICAL RESEARCH

Exposure assessment is considered to be the Achilles heel of air pollutant epidemiology (Jerrett, 2013, personal communication). In many studies exposure is estimated using simple methodologies not completely or not accurately reflecting real exposure. As a result, exposure misclassification will arise and health effects may not be identified, the effect will be smaller than in reality or an erroneous effect will be detected (Özkaynak et al., 2013; Setton et al., 2011; Sheppard et al., 2012). Improving exposure estimates, e.g. by taking into account air pollution surfaces, time-activity patterns or direct personal measurements, is identified as the way forward in large epidemiological studies (Fenske, 2010). This is especially the case for many traffic related pollutants such as BC that are characterized by a high spatiotemporal heterogeneity.

Personal measurements, as shown in chapter 3, are one way of estimating personal exposure more accurately. Personal measurements are realistic for short term exposures, but not necessarily for longer term exposures or for historic exposures. The modeling framework AB²C developed in chapter 4 can be

used to predict exposure of a large group of individuals or for a subset of the population using only very limited input-data. For every person a range of exposures will be predicted based on the changing time-activity pattern of an individual. Not only long term exposures can be calculated or the fraction of a population exposed to levels above a certain threshold, but also the frequency of short term exposures above a certain level can be determined. The latter may bring new opportunities to epidemiologists: studying the effect of frequently repeated but short exposure peaks on long term exposure and health.
The growing field of spatial epidemiology can benefit from the new GIS methods presented in this PhD (Jerrett et al., 2010): both the personal measurements using GPS, the time-location-activity diaries from the FEATHERS model integrated in AB²C, and the hourly LUR models, are concepts that can readily be incorporated in epidemiological studies.

It is yet to be investigated whether the improved exposure estimates developed in this PhD result in a gain for epidemiologists, and how large that gain is (Szpiro et al., 2011). As described earlier, the advantage of using an AB²C-like framework to estimate personal exposure to PM_{10} will be very limited, on the contrary using this framework will introduce errors not present when using raw measurement data on fixed monitors. Pollutants that are mainly traffic-related will be aided by the use of a more detailed exposure assessment using personal monitoring or exposure models. In that way, this study will contribute to the design of future health studies that seek to study the relationship between observed health effects and traffic.

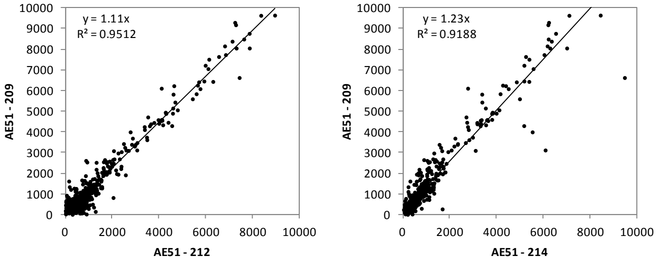
A. APPENDIX

All appendices can be accessed online as supplemental material associated with the respective publications.

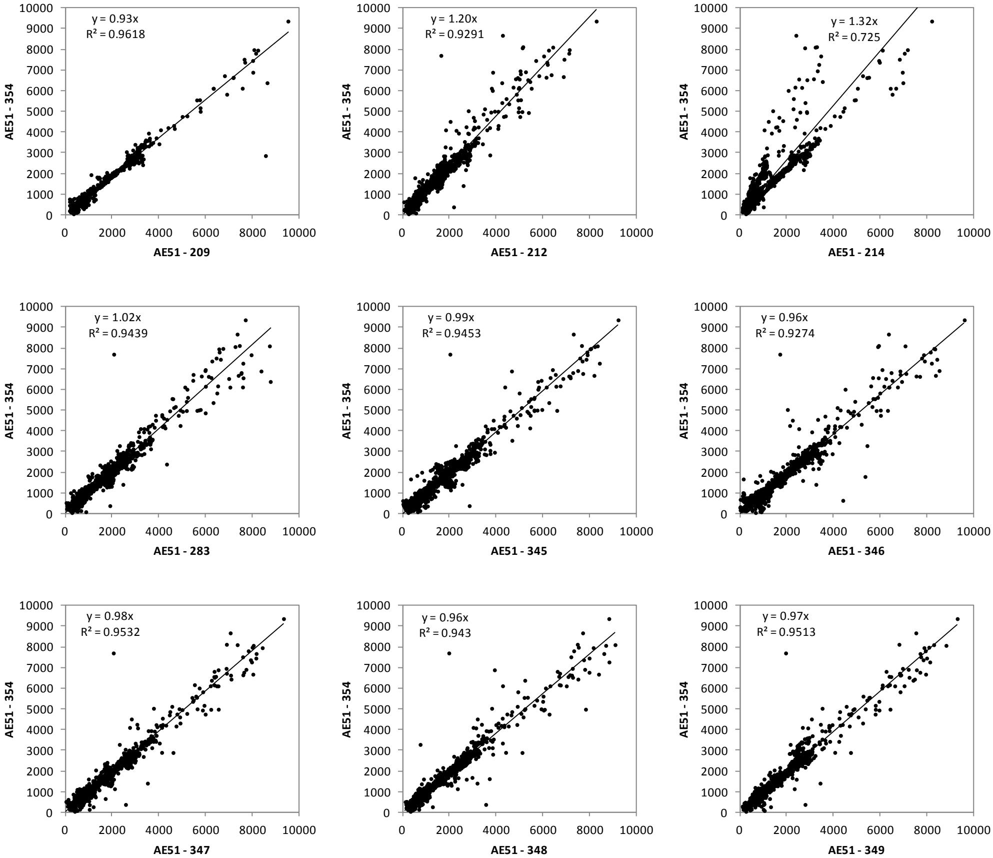
A.1 SUPPLEMENTAL MATERIAL CHAPTER 3.2

Intercomparison of 13 aethalometers model AE51

Intercomparison summer (post-campaign comparison):



Intercomparison winter (pre- and post-campaign comparison)



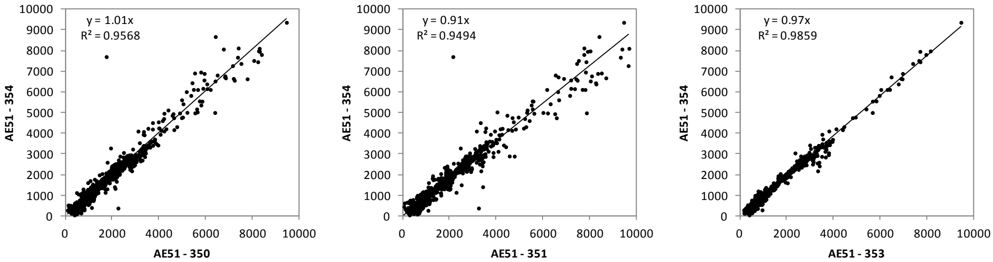


FIGURE A1: Results of the intercomparison of 13 aethalometers. Device 214 was excluded from the winter field campaign because of deviating values during this intercomparison. The slope of the regression function is used to correct personal measurements during the field campaign. Pearson's r is consistently high (0.96-0.99), except for device 214 in winter.

Correction for variation in background concentrations

Due to the limited availability of measurement devices, we were obliged to spread the measurements over several weeks. To be able to compare concentrations measured in different microenvironments directly, without the influence of varying background concentrations, we calculated corrected or 'rescaled' concentrations. Corrections are based on measurements from a fixed BC reference monitor operated by the Flemish Environment Agency on a suburban background location (station 40AL01 – Antwerpen Linkeroever). At this location local traffic has only a limited impact on BC concentrations; the observed temporal variation is representative for the study area.

It is an unsolved question whether the day-to-day variations of the background monitor are additive or multiplicative. As an example, Zuurbier et al. (2010) corrected air pollution concentrations for absolute differences between the mean background concentrations during all sampling days and the background concentration of each sampling day. She followed previous studies (Cyrys et al., 2003). Hoek et al. (2002) compared the additive and the multiplicative method and for their study areas, the additive method gave slightly better results.
For chapter 3.2, we did a sensitivity analysis using four different methodologies, including unadjusted concentrations. The impact of type of correction on FIGURE 18, FIGURES A5 and A6 is calculated. The results are presented in TABLE A1a-c and FIGURE A3a-c. The main conclusion is that corrections, independent of the rescaling method, are always smaller than differences between different activities and between different microenvironments. As most important results:

- The multiplication method overcorrects high peak exposures, originating from local sources. Consequently the average concentration will be overestimated.
- Median concentrations are not influenced by the rescaling method.
- The additive method is introducing negative values, e.g. P5 is more often negative in the additive method compared to all other methods.
- The method used in chapter 3.2, i.e. a combination of the additive and multiplicative method is relying on the additive method for higher values and on the multiplicative method for lower values. The impact of this rescaling

on FIGURE 18, FIGURES A5 and A6 is in between the impact of the 'multiplicative only' and 'additive only' method.

- When averaging results over the whole study period, the 'unadjusted concentrations' option is very similar to any of the possible rescaling methods; although this hides the impact on individual measurements.

Based on the results of the sensitivity analysis, it was decided to use two different rescaling formulas, dependent on the difference between the daily average concentration measured at the reference station and the 5-min personal exposure of the volunteer. If a personal measurement is higher than the daily average concentration at the reference station (personal measurement > refsite_DailyAverage), the absolute difference between yearly and daily average concentrations at the reference site was used as correction. We used the absolute difference because, when using a multiplication, the part of the personal exposure originating from local sources would also be affected, which is counterintuitive and unwanted. In all other cases (personal measurement < refsite_DailyAverage), we used the ratio of yearly and daily average concentrations at the reference site. For these lower concentrations, we worked with a ratio rather than with the difference between daily and yearly averages at the reference station, mainly to prevent corrected concentrations becoming negative. In this case we assume no local sources of BC, and thus the personal measurement in its entirety is corrected for variation observed at the reference site. Whether this correction works in indoor microenvironments is an open question, but we observed a high correlation between indoor and outdoor concentrations at the residential location of the participants (r=0.81).
The rescaling algorithm is programmed and calculated in SAS 9.2 (schematic representation in FIGURE A2).

Personal measurement > refsite_DailyAverage	Personal measurement < refsite_DailyAverage
Personal measurement_corr = Personal measurement + (refsite_YearlyAverage – refsite_DailyAverage)	Personal measurement_corr = Personal measurement * (refsite_YearlyAverage / refsite_DailyAverage)

FIGURE A2: Schematic representation of the formulas used for the rescaling of personal measurements

TABLE A1a: Overview of the impact of the rescaling method on concentrations in different microenvironments. Three methods are presented: the correction applied in chapter 3.2 and 3.3 (combination of multiplicative and additive method), multiplicative method only or additive correction. Results are compared to uncorrected measurements.

Chapter 3.2 + 3.3	In transport	Other	Family / Friends	Home	Work / School
P5	490	-69	351	299	78
Q1	1895	836	901	636	455
Median	3507	1830	1324	1001	873
Q3	6298	3080	1786	1604	1425
P95	15444	9601	3976	2805	2786
Avg	4976	2840	1565	1246	1068
Count	7039	4742	2259	72406	17298
No correction	In transport	Other	Family / Friends	Home	Work / School
P5	409	-53	203	220	95
Q1	1792	687	645	554	443
Median	3596	1464	1032	971	837
Q3	6529	3021	1747	1597	1417
P95	15672	9284	4426	3142	2847
Avg	5096	2714	1446	1255	1087
Count	7039	4742	2259	72406	17298
Only multiplicative	In transport	Other	Family / Friends	Home	Work / School
P5	490	-69	351	299	78
Q1	1900	836	901	636	455
Median	3636	1962	1324	1001	873
Q3	7104	4178	1977	1604	1425
P95	19515	15964	5251	3326	3165
Avg	5999	4079	2315	1369	1126
Count	7039	4742	2259	72406	17298
Only additive	In transport	Other	Family / Friends	Home	Work / School
P5	221	-492	57	-702	-985
Q1	1895	913	884	463	210
Median	3507	1830	1422	1118	893
Q3	6298	3080	1786	1610	1480
P95	15444	9601	3976	2805	2786
Avg	4974	2857	1560	1118	895
Count	7039	4742	2259	72406	17298

TABLE A1b: Overview of the impact of the rescaling method on concentrations during different activities. Three methods are presented: the correction applied in chapter 3.2 and 3.3 (combination of multiplicative and additive method), multiplicative method only or additive correction. Results are compared to uncorrected measurements.

Chapter 3.2 + 3.3	In transport	Go for a ride	Social and leisure	Shopping	Other	Home-based activities	Work	Sleep
P5	548	-241	190	204	148	293	84	301
Q1	2013	908	833	952	675	710	483	583
Median	3685	1680	1366	1617	1313	1132	917	892
Q3	6511	2827	2304	2918	2132	1754	1441	1405
P95	15569	14341	8723	7694	5443	3134	2679	2492
Avg	5132	2855	2445	2540	1829	1360	1077	1090
Count	6560	481	5878	1243	2992	31558	18204	36828
No correction	In transport	Go for a ride	Social and leisure	Shopping	Other	Home-based activities	Work	Sleep
P5	508	-127	87	108	116	232	103	211
Q1	1993	710	655	1026	590	621	460	497
Median	3771	1526	1149	1837	1040	1059	855	879
Q3	6735	2921	2182	3204	2028	1734	1423	1468
P95	15770	14124	8496	8261	5497	3459	2831	2756
Avg	5262	2823	2376	2721	1713	1353	1087	1108
Count	6560	481	5878	1243	2992	31558	18204	36828
Only multiplicative	In transport	Go for a ride	Social and leisure	Shopping	Other	Home-based activities	Work	Sleep
P5	548	-241	190	204	148	293	84	301
Q1	2012	908	833	952	675	710	483	583
Median	3788	1726	1366	1617	1313	1132	917	892
Q3	7363	3629	2649	3463	2426	1840	1441	1405
P95	19809	11245	13323	9299	8100	4027	3137	2778
Avg	6201	3251	3303	2925	2278	1591	1136	1151
Count	6560	481	5878	1243	2992	31558	18204	36828
Only additive	In transport	Go for a ride	Social and leisure	Shopping	Other	Home-based activities	Work	Sleep
P5	350	-336	-495	-629	-416	-697	-971	-712
Q1	2013	1021	880	744	623	527	240	414
Median	3685	1680	1455	1616	1417	1234	924	1022
Q3	6511	2827	2304	2918	2132	1754	1500	1480
P95	15569	14341	8723	7694	5443	3134	2679	2492
Avg	5127	2881	2426	2418	1769	1236	906	962
Count	6560	481	5878	1243	2992	31558	18204	36828

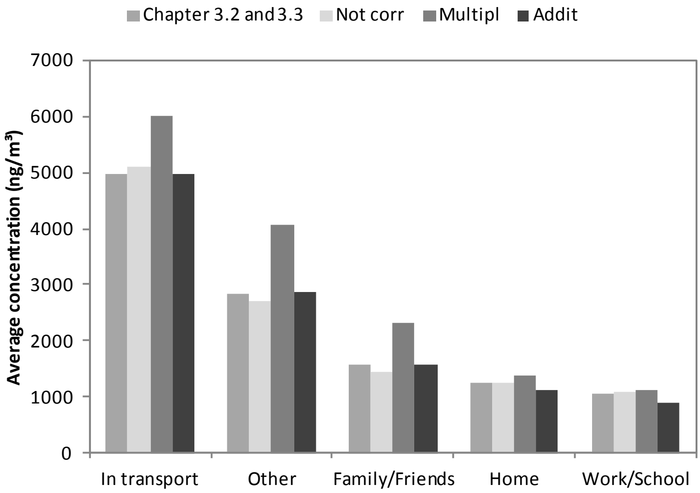
TABLE A1c: Overview of the impact of the rescaling method on concentrations in different transport modes. Three methods are presented: the correction applied in chapter 3.2 and 3.3 (combination of multiplicative and additive method), multiplicative method only or additive correction. Results are compared to uncorrected measurements.

Chapter 3.2 + 3.3	Car driver	Car passen-ger	Bike	On foot	Train	Light rail / metro	Bus
P5	713	991	187	25	460	1436	1411
Q1	2530	2483	1625	1313	1263	2987	2893
Median	4530	4350	2858	2393	2062	4913	4922
Q3	8327	7570	4732	4084	3181	7319	7692
P95	19161	13595	9969	9139	5415	8814	17423
Avg	6432	5583	3555	3175	2394	5066	6575
Count	3190	645	1167	1161	614	72	190

No correction	Car driver	Car passen-ger	Bike	On foot	Train	Light rail / metro	Bus
P5	690	913	167	12	480	1487	1922
Q1	2610	2536	1309	1122	1117	3205	3089
Median	4893	4528	2588	2539	2021	5404	5285
Q3	8643	7799	4787	4215	3297	8100	8345
P95	19152	13759	9786	9146	5363	9621	17651
Avg	6646	5675	3479	3236	2436	5556	6844
Count	3190	645	1167	1161	614	72	190

Only multipli-cative	Car driver	Car passen-ger	Bike	On foot	Train	Light rail / metro	Bus
P5	713	991	187	25	460	1436	1411
Q1	2478	2550	1624	1313	1263	2808	3026
Median	4616	4801	3045	2553	2129	4291	5437
Q3	9219	9102	5739	4903	3383	5873	9336
P95	23744	24107	14786	11526	6725	11978	19270
AVG	7461	7598	4884	3755	2875	4867	7165
Count	3190	645	1167	1161	614	72	190

Only additive	Car driver	Car passen-ger	Bike	On foot	Train	Light rail / metro	Bus
P5	518	925	-78	-285	83	1247	1214
Q1	2530	2483	1625	1348	1251	2987	2893
Median	4530	4350	2858	2393	2062	4913	4922
Q3	8327	7570	4732	4084	3181	7319	7692
P95	19161	13595	9969	9139	5415	8814	17423
AVG	6422	5561	3586	3184	2377	5013	6549
Count	3190	645	1167	1161	614	72	190



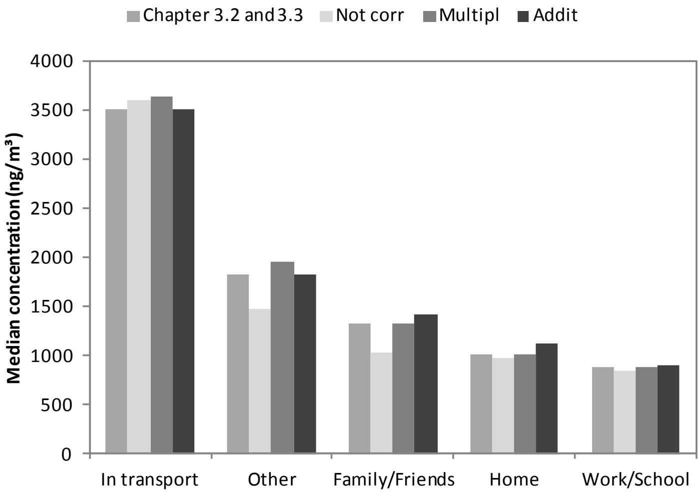
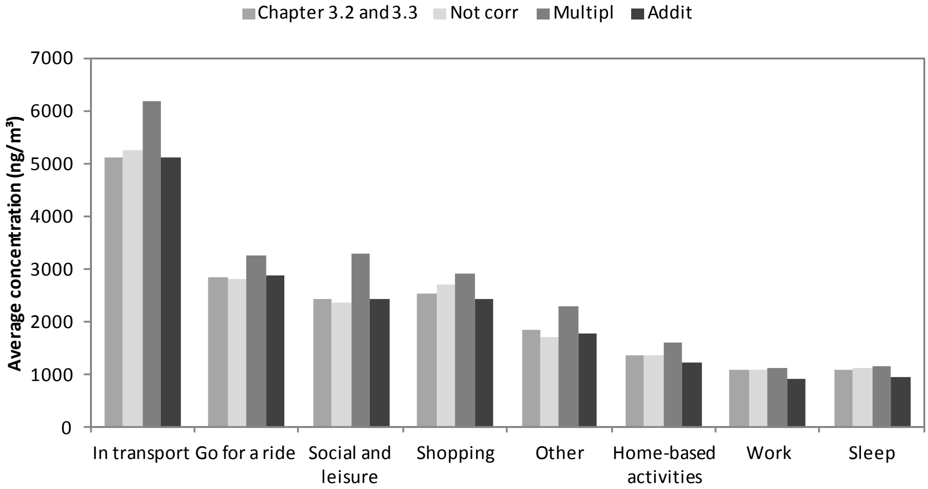


FIGURE A3a: Overview of the impact of the rescaling method on concentrations in different microenvironments. Average and median concentrations from TABLE A1a are shown.



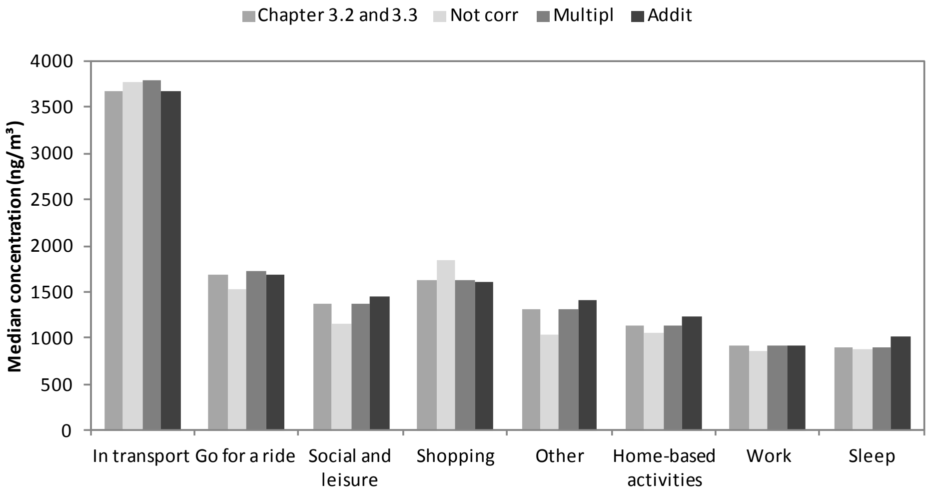
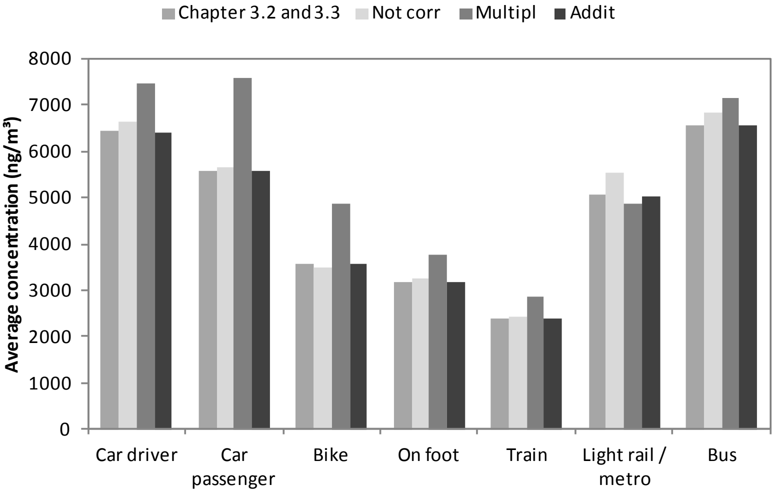


FIGURE A3b: Overview of the impact of the rescaling method on concentrations during different activities. Average and median concentrations from TABLE A1b are shown.



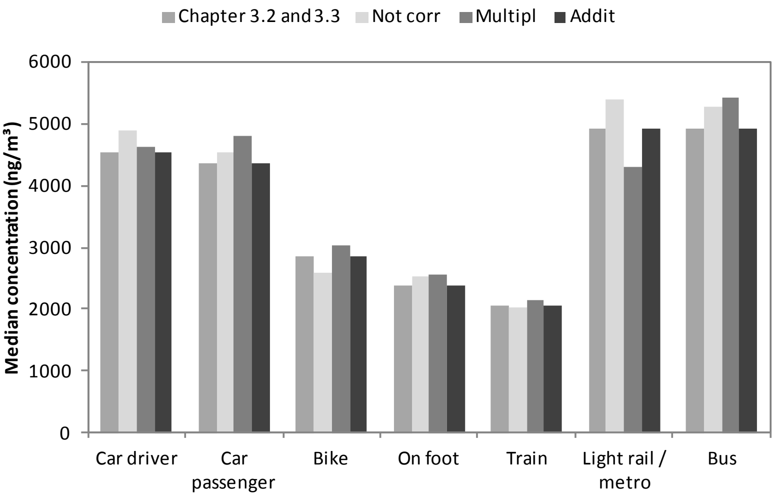


FIGURE A3c: Overview of the impact of the rescaling method on concentrations in different transport modes. Average and median concentrations from TABLE A1c are shown.

Minute volume assumptions

Inhalation rates are based on Allan & Richardson (1998) and Int Panis et al. (2010). Activities and transport modes are linked to activity levels in an ad hoc manner.

TABLE A2: Summary of minute volume assumptions (l/min). Numbers based on Allen & Richardson (1998); except for the rows with the asterisk which are based on Int Panis et al. (2010).

Activity	Male adults	Female adults
Sleep	8.3	7.5
Home-based activities / Eat / Education	10.5	12.5
Work / Social / Other / Service related activities / Bring/get goods/people	16.1	13.0
Leisure / Daily shopping / Non-daily shopping	30.2	23.2
On foot	49.2	39.8
Train / Bus / Light rail / Metro	16.1	13.0
Car driver / Car passenger / Taxi *	13.4	11.3
Bike *	59.1	46.2

Study area

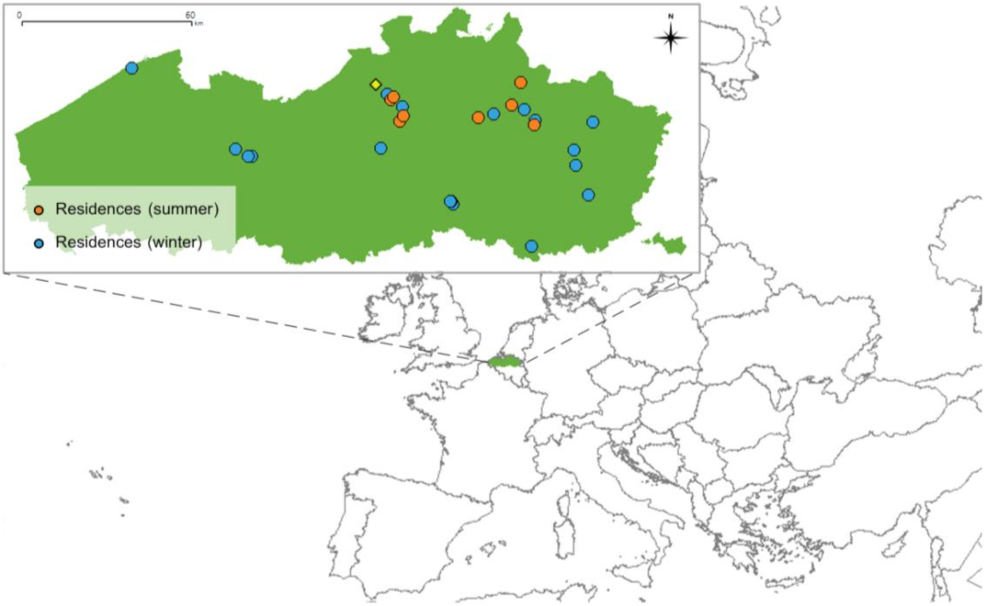


FIGURE A4: Study area with residential location of participating couples. The yellow diamond marks the location of the suburban background monitor we used to correct for non-simultaneous measurements.

Vehicle characteristics

For the analysis we relied on a large dataset of over 1000 car trips. Participating households owned 45 cars in total, an average of almost 1.5 car per household. Almost half of the cars (21 cars) did not meet the Euro 4 or Euro 5 emission standards (based on year of manufacture), and 31 cars were diesel; information that was drawn from a questionnaire filled in by all volunteers.

TABLE A3: Vehicle characteristics and car ownership per household participating in the study

Season	Household Number	# cars (total per household)	# diesel cars	Year of manufacture car 1	Year of manufacture car 2	Year of manufacture car 3
1	HH1	2	2	2005 or older	2006 or newer	
1	HH2	1	1	2005 or older		
1	HH3	1	1	2005 or older		
1	HH4	1	1	2005 or older		
1	HH5	2	2	2005 or older	2006 or newer	
1	HH6	0	0			
1	HH7	2	2	2006 or newer	2006 or newer	
1	HH8	2	2	2005 or older	2006 or newer	
2	HH1	2	2	2005 or older	2006 or newer	
2	HH2	1	1	2005 or older		
2	HH4	1	1	2005 or older		
2	HH8	2	2	2005 or older	2006 or newer	
2	HH9	2	2	2006 or newer	2006 or newer	
2	HH10	1	0	2005 or older		
2	HH11	1	1	2005 or older		
2	HH12	1	1	2006 or newer		
2	HH13	2	2	2006 or newer	2006 or newer	
2	HH14	1	1	2005 or older		
2	HH15	1	0	2005 or older		
2	HH16	1	1	2005 or older		
2	HH17	0	0			
2	HH18	2	1	2005 or older	2006 or newer	
2	HH19	1	1	2006 or newer		
2	HH20	3	3	2005 or older	2005 or older	2006 or newer
2	HH21	1	1	2006 or newer		
2	HH22	2	1	2005 or older	2006 or newer	
2	HH23	1	1	2006 or newer		
2	HH24	2	1	2005 or older	2006 or newer	
2	HH25	3	3	2006 or newer	2006 or newer	2006 or newer
2	HH26	2	2	2005 or older	2006 or newer	
2	HH27	1	1	2006 or newer		

Over 60% of all private cars in Belgium are diesel (NIS, 2010); this number is remarkably higher than in the neighboring countries (e.g. in the Netherlands, 17% of all passenger cars have diesel engines (CBS, 2008)). Because diesel is the cheaper fuel, diesel cars travel, on average, more kilometers per year. In addition, a large part of the traffic on the main roads is heavy traffic (caused by the role of Belgium as a transit country, and the vicinity of several large ports).

Concentrations measured in different microenvironments and during different activities

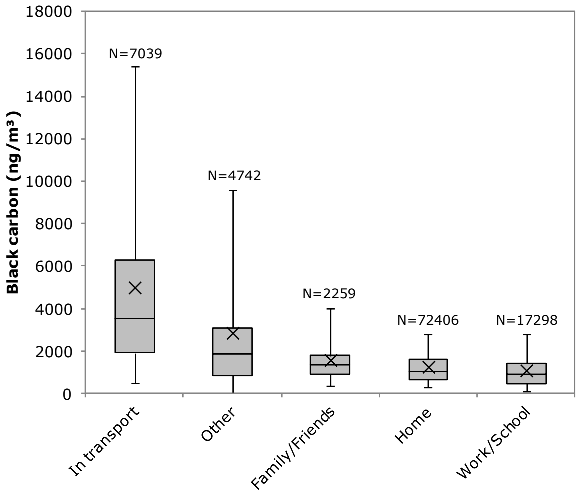


FIGURE A5: Concentrations measured in different microenvironments. The 'N=' indicates the number of 5-minute observations used to calculate the average and the percentiles. Represented are P5, 1st quartile, median, 3rd quartile and P95. The cross marks the mean value.

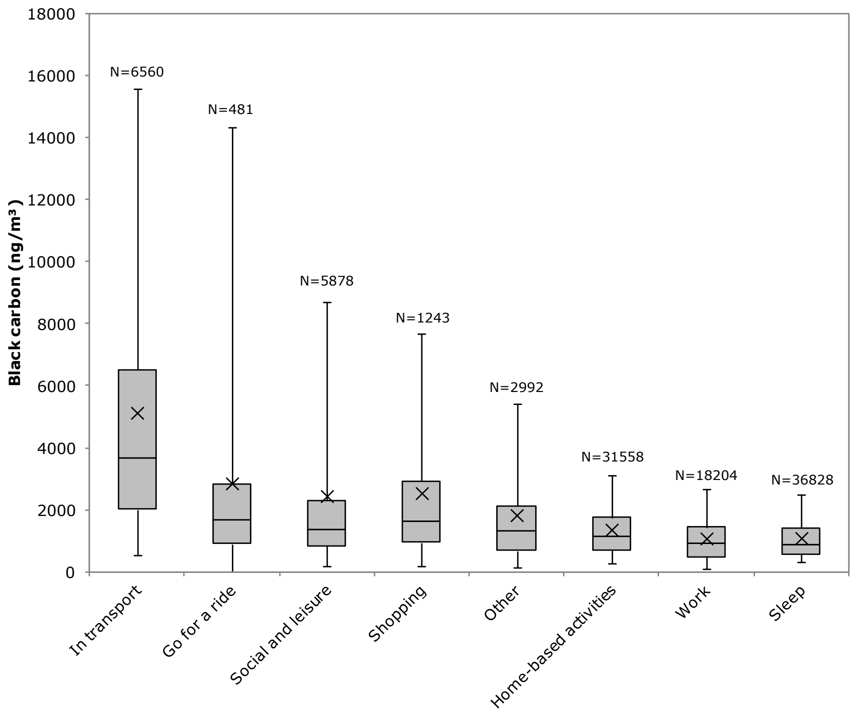


FIGURE A6: Concentrations measured during different activities. The 'N=' indicates the number of 5-minute observations used to calculate the average and the percentiles. Represented are P5, 1st quartile, median, 3rd quartile and P95. The cross marks the mean value.

Travel time versus weeklong average exposure

Because multiple factors influence exposure in transport, it is not straightforward to relate travel time to integrated personal exposure. We show that average exposure in transport can be highly variable between individuals depending on the transport modes used, and the timing of trips (time-of-day, day of the week). 'Time in transport' and 'Car travel time' are not good predictors of weeklong personal exposure or dose, as illustrated in the correlation plots in FIGURE 21 and FIGURE A7.

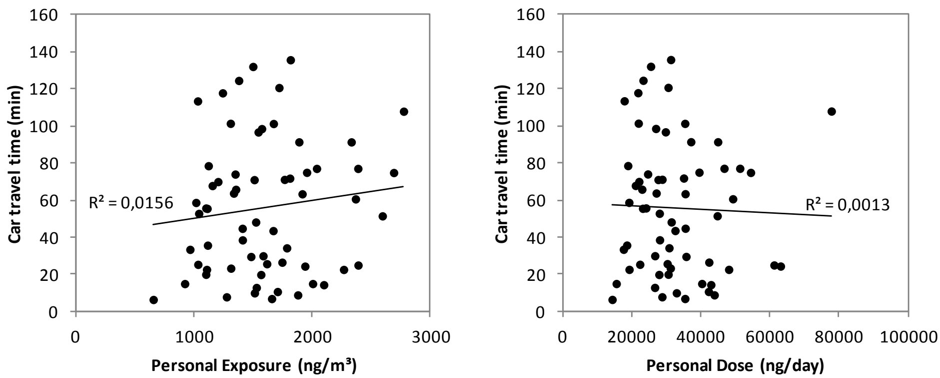


FIGURE A7: Correlation between personal exposure and car travel time; and correlation between personal dose and car travel time. Each mark represents one of 62 volunteers. Car travel time is defined as the time, in minutes, spent as car driver or car passenger.

In contrast with the poor correlation between travel time and personal exposure, our results do reveal a better correlation between average exposure in transport and average personal exposure (FIGURE A8). This is in line with the finding that 21% of exposure is attributable to exposure during travel. Because we know that trips are responsible for 30% of daily inhaled dose, the relationship between average dose in transport and personal dose is expected to be a little better: the explained variance is 36%.

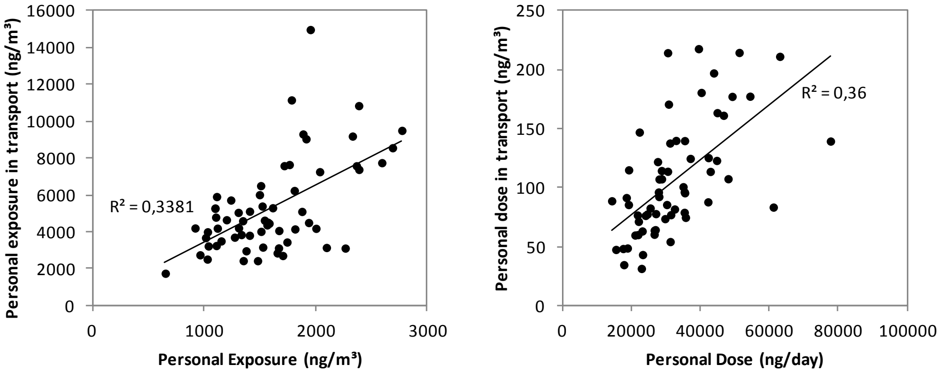


FIGURE A8: Correlation between personal exposure and exposure in transport (left) and personal dose and dose in transport (right). Each mark represents one of 62 volunteers.

The predictability of personal exposure by residential outdoor concentrations is also explored, resulting in an R^2 of 0.32, based on a subset of 25 couples with fixed outdoor BC measurements at their residence. These BC measurements were done simultaneously with the personal measurements, using the same equipment and standards as explained in the 'Materials and methods'-section (chapter 3.2.2).

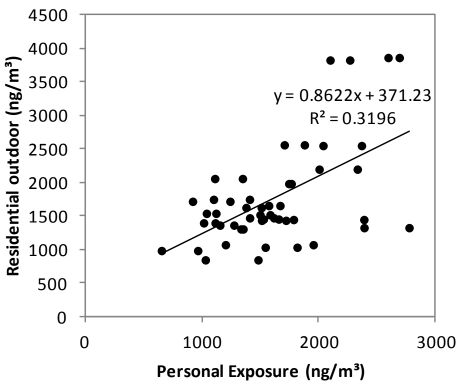


FIGURE A9: Correlation between personal exposure and BC concentrations at the residence of 50 participants.

TABLE A4: Exposure-ratios and dose-ratios by transport mode, separated by rush hour and non-rush hour

	All data		Morning rush hour (7-10 a.m.)		Evening rush hour (4-7 p.m.)		Non-rush hour	
	Exp-ratios	Dose-ratios	Exp-ratios	Dose-ratios	Exp-ratios	Dose-ratios	Exp-ratios	Dose-ratios
Automobile: bicycle	1.77	0.41	1.94	0.46	2.15	0.52	1.47	0.32
Automobile: foot	2.00	0.56	2.19	0.61	2.32	0.65	1.65	0.46
Automobile: bus	0.96	0.82	0.83	0.73	1.08	0.94	1.06	0.89
Automobile: train	2.63	2.16	3.06	2.57	2.95	2.38	2.32	1.92

TABLE A5: Exposure-ratios and dose-ratios by transport mode, separated by weekday and weekend

	All data		Weekday		Weekend	
	Exposure-ratios	Dose-ratios	Exposure-ratios	Dose-ratios	Exposure-ratios	Dose-ratios
Automobile: bicycle	1.77	0.41	1.79	0.41	1.83	0.42
Automobile: foot	2.00	0.56	1.76	0.49	2.73	0.77
Automobile: bus	0.96	0.82	0.99	0.84	0.86	0.82
Automobile: train	2.63	2.16	2.76	2.25	2.43	2.29

In-car concentrations according to trip motive

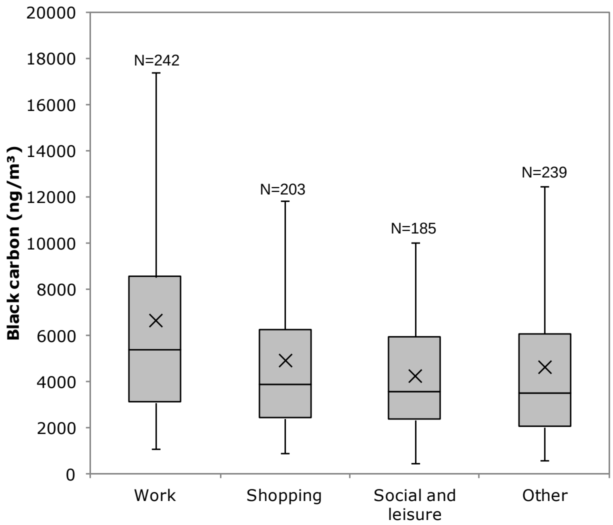


FIGURE A10: Average in-car BC concentrations (ng/m³) according to trip motive. The 'N=' indicates the number of trips used to calculate the average and the percentiles. Represented are P5, 1st quartile, median, 3rd quartile and P95. The cross marks the average.

A.2 SUPPLEMENTAL MATERIAL CHAPTER 3.3

TABLE A6: Description of road network data

Description of dataset	Flanders road network with linked peak hour traffic intensities (see FIGURE A11 for a visual representation).
Name of dataset	Multimodaal model Vlaanderen (MMM2007) (Flemish multimodal traffic model)
Type of data	Vector
Accuracy/resolution dataset	Approximately 40% of total road length is included in this dataset, including all the main roads. No traffic is assigned to 3% of included segments.
Completeness	Covers whole of Flanders (Northern Belgium) excluding Brussels
Coordinate system	Lambert-72 (Lambert Conformal Conic)
Year(s) for which data are available	2007
Source of the data	Flemish Traffic Control Center (Vlaams Verkeerscentrum)
Available Data fields	Geocoded location, Road type, Urbanization, Free flow speed, Peak traffic flow
Remarks	Only evening peak hour intensities for an average workday were available. For light traffic, these were multiplied by 12.02 to obtain daily workday traffic intensity and by 0.96 to obtain 24-h averages over entire weeks, following the standards of the Belgian CAR manual*. For heavy traffic a similar methodology was used: peak hour intensities were multiplied by 18.55 to obtain daily traffic intensities, and by 0.80 to obtain 24-h averages over entire weeks. Total traffic is calculated as the sum of light and heavy traffic.

* http://www.tmleuven.be/project/car/Handleiding_CAR-Vlaanderen_v2.0.pdf. (In Dutch)

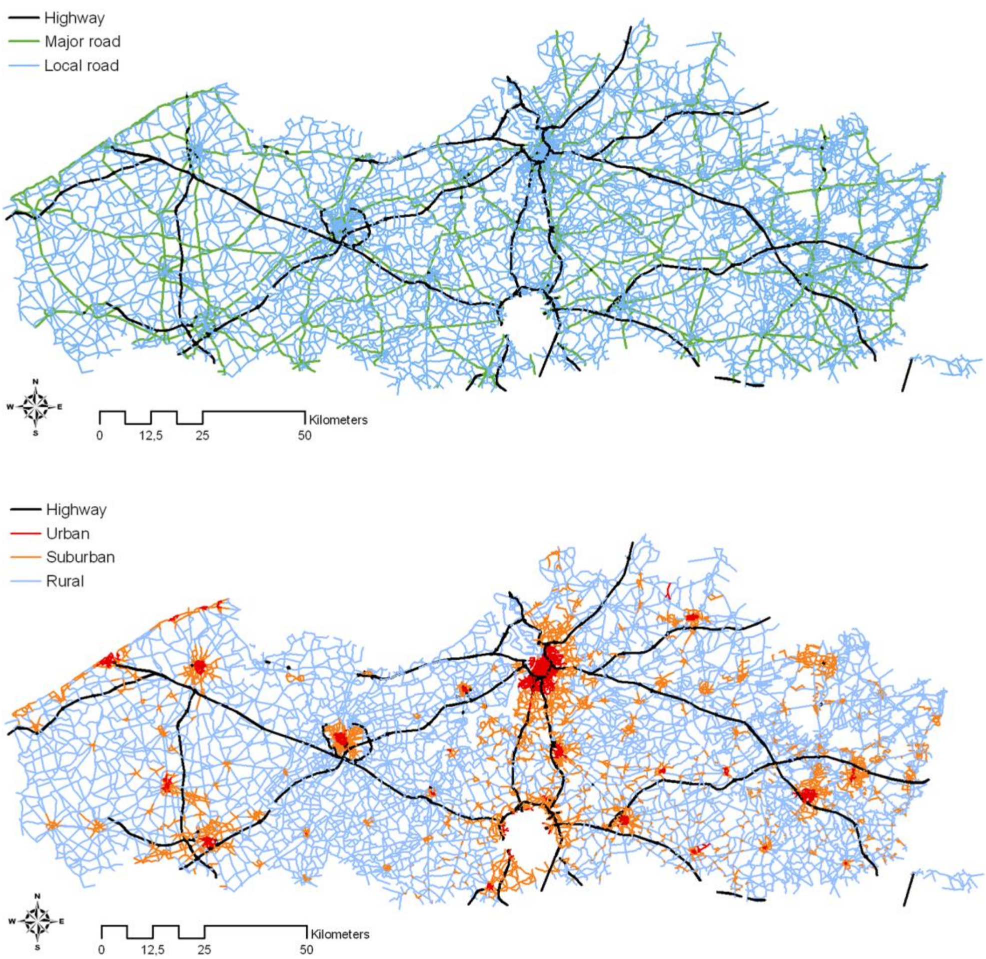


FIGURE A11: Different visual representations of the Flemish multimodal traffic model map used to link to geocoded BC-measurements.

The map contains all highways and major roads but only 40% of the total length of road segments. Trajectories of roads are mostly geographically accurate but sometimes contain linear simplifications of curving segments. The absence of many small roads (such as agricultural roads, parallel service roads, unpaved roads and dedicated off-road bike paths without any motorized traffic) actually reduces the risk of misclassification.

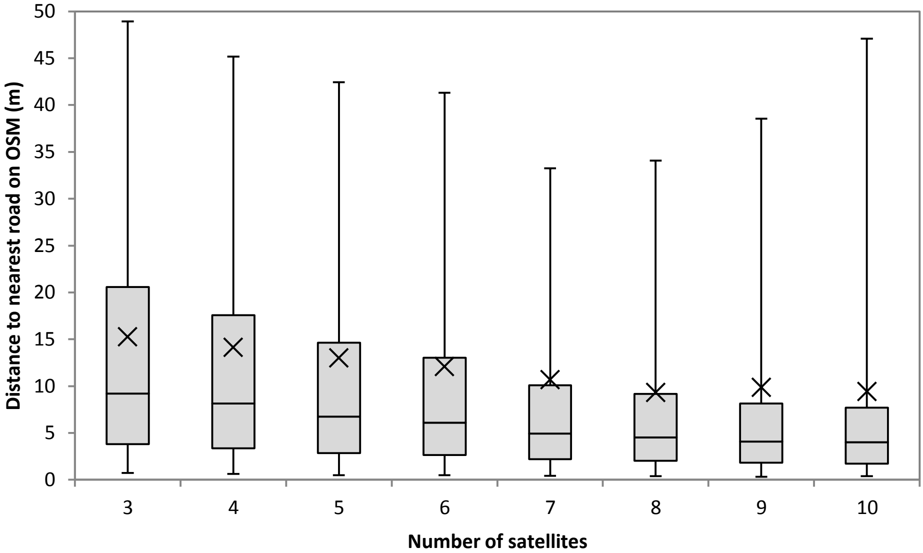


Figure A12: Distance of geocoded BC observations to the nearest road (using the OpenStreetMap network) as a function of the number of satellites in view. Boxplots show 5th, 25th, 75th, 95th percentiles, median and mean (×). Locations based on 3 and 4 satellites were excluded from the analysis.

Using a more complete OpenStreetMap (OSM) dataset would result in more observations being unequivocally linked to a road. Although the OSM contains more road segments, it lacks the associated traffic data needed for our model hence it could not be used.

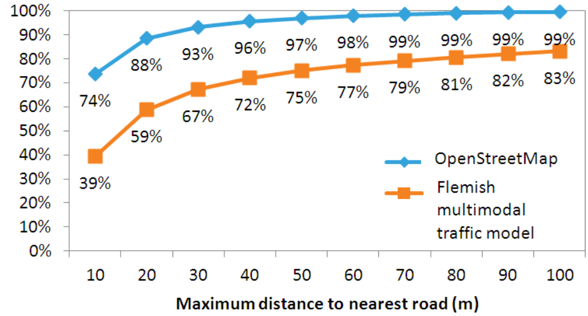


Figure A13: When linked to the Flemish multimodal traffic model map, 67% of BC observations are located less than 30 m from a road. Distances shown include both errors in the GPS location and geographical accuracy of the map.

We have arbitrarily decided to include only observations when a mapped road segment was found within 30 meter. Also if both the location of the BC measurement and the location of the road are correct (e.g. when entering a driveway or parking lot), the BC concentration due to the road would have leveled off substantially (Zhu et al., 2002) which would hamper attempts to model effects of road characteristics on BC concentrations. The main effect of this selection will be from the omission of observations on very small roads with low BC concentrations (FIGURE A14). Observations of high BC-concentrations are conserved because all major roads are present in the dataset.

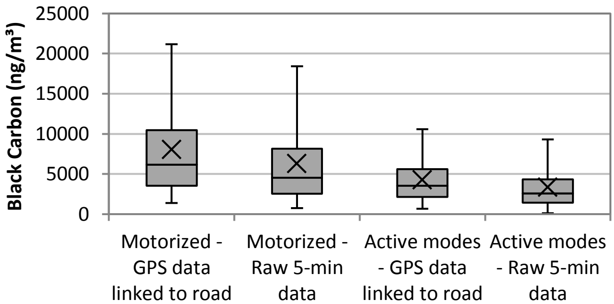


Figure A14: Distribution of raw 5-min data compared to the observations left after assigning waypoints to a road (max distance 30m) – Boxplot showing 5th, 25th, 75th, 95th percentiles, median and mean (×).

TABLE A7: Daily traffic intensities are translated into hourly 'instantaneous' traffic intensities based on the CAR-table presenting percentages of cars passing each hour of the day. Separate factors are specified for weekdays, Saturdays and Sundays (Jonkers and Vanhove, 2010).

All traffic	Monday-Friday %	Saturday %	Sunday %
0u-1u	0.42	1.47	2.18
1u-2u	0.26	0.97	1.59
2u-3u	0.16	0.66	1.12
3u-4u	0.24	0.53	0.86
4u-5u	1.02	0.53	0.69
5u-6u	2.52	0.84	0.77
6u-7u	3.78	1.28	1.02
7u-8u	8.00	2.16	1.45
8u-9u	7.39	4.06	2.60
9u-10u	5.27	5.96	4.35
10u-11u	5.41	7.08	5.81
11u-12u	5.22	7.13	6.91
12u-13u	5.56	6.48	6.36
13u-14u	6.39	6.88	6.00
14u-15u	5.94	7.69	7.13
15u-16u	6.05	7.50	7.40
16u-17u	8.14	7.52	7.84
17u-18u	8.32	7.70	8.51
18u-19u	6.41	7.12	8.17
19u-20u	4.11	5.65	6.79
20u-21u	3.11	3.57	5.16
21u-22u	3.02	2.47	3.54
22u-23u	2.13	2.47	2.38
23u-24u	1.11	2.28	1.36

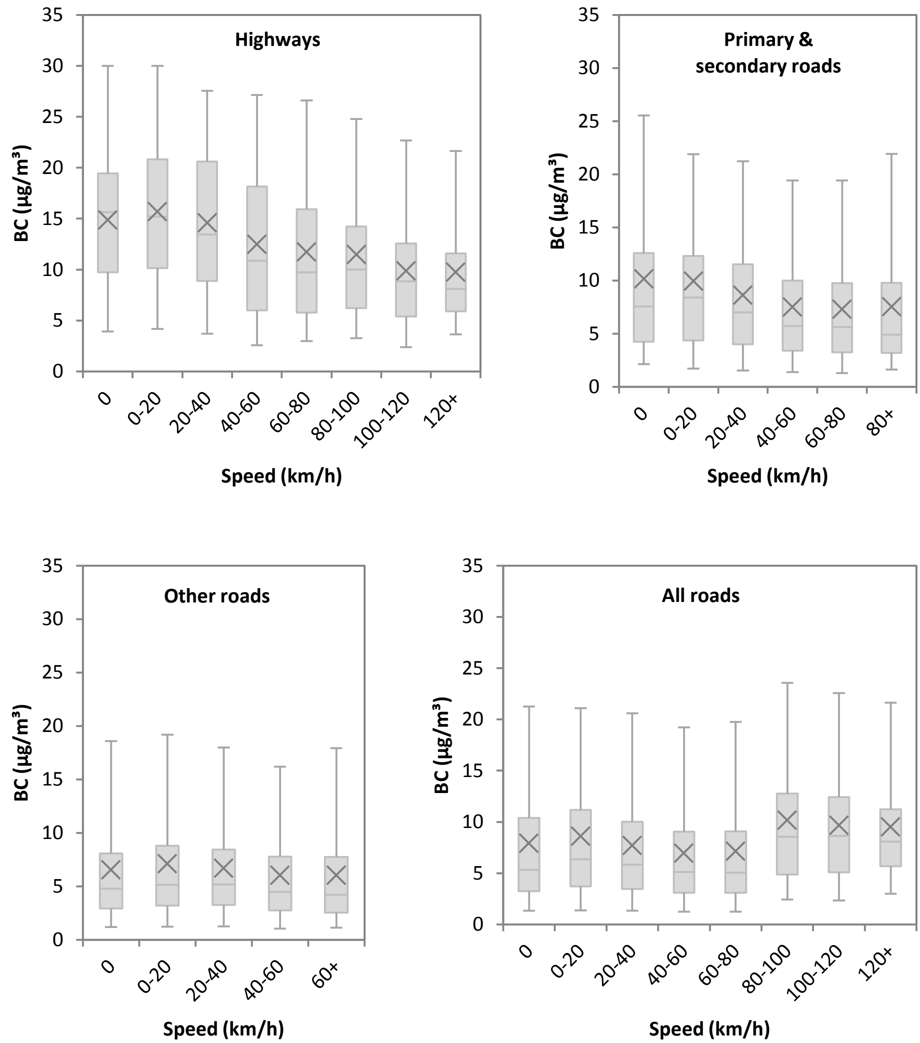


Figure A15: BC as a function of travel speed (GPS based) in cars. Boxplot showing 5th, 25th, 75th, 95th percentiles, median and mean (×).

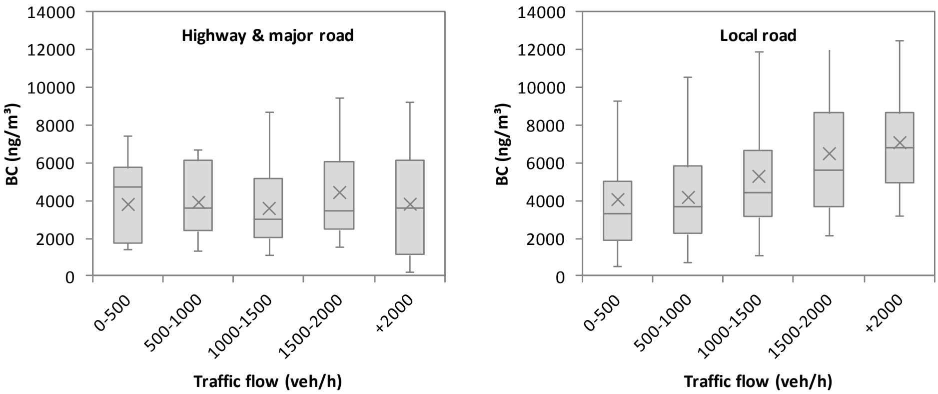


FIGURE A16: BC as a function of traffic intensity, and according to road type for active travelers. Boxplot showing 5th, 25th, 75th, 95th percentiles, median and mean (×).

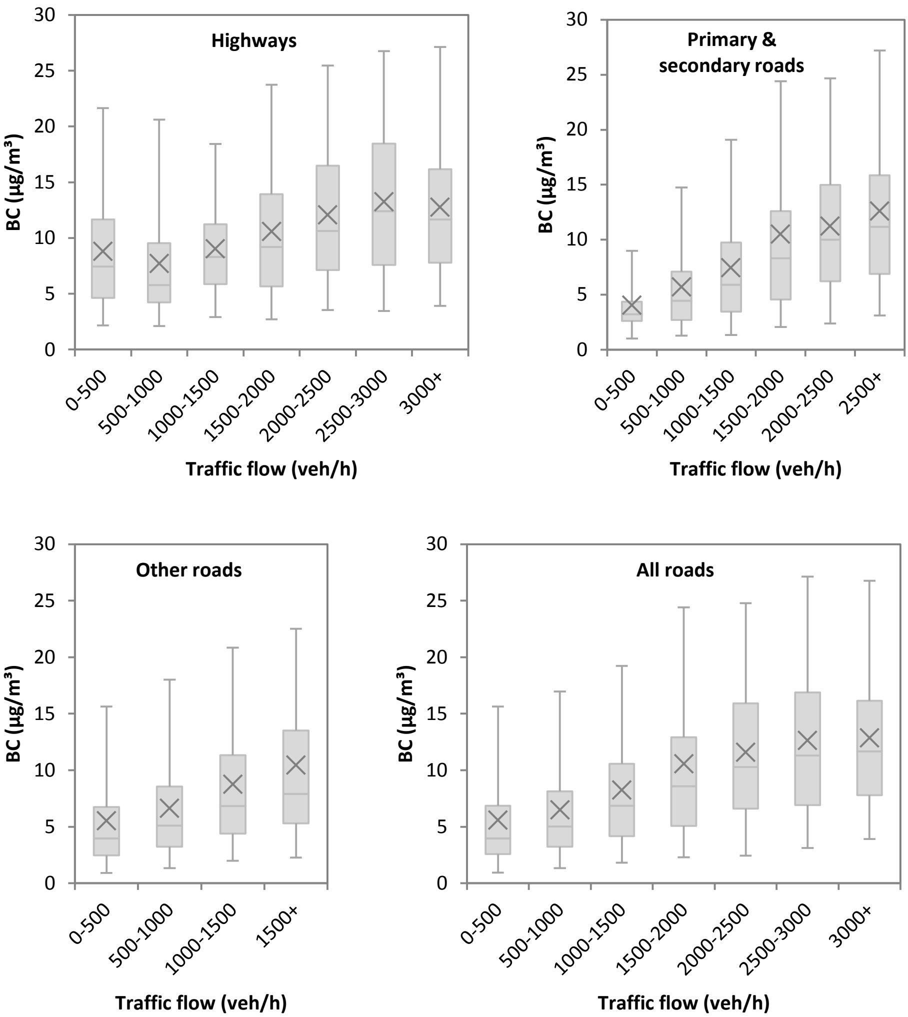


Figure A17: In-vehicle BC as a function of traffic flow. Boxplot showing 5th, 25th, 75th, 95th percentiles, median and mean (×).

TABLE A8: Regression model for BC exposure in motorized transport (R^2=0.17)

Variable	DF	Parameter Estimate	Standard Error	t Value	Pr > |t|
Intercept	1	2947	23.9	123.5	<.0001
Traffic intensity (veh/h)	1	1.549	0.009	163.6	<.0001
Peak	1	3289	22.7	145.0	<.0001
Off-peak	1	1189	24.1	49.3	<.0001
Highway	1	2309	26.5	87.3	<.0001
Urban	1	2853	27.3	104.5	<.0001
Suburban	1	1167	21.9	53.2	<.0001

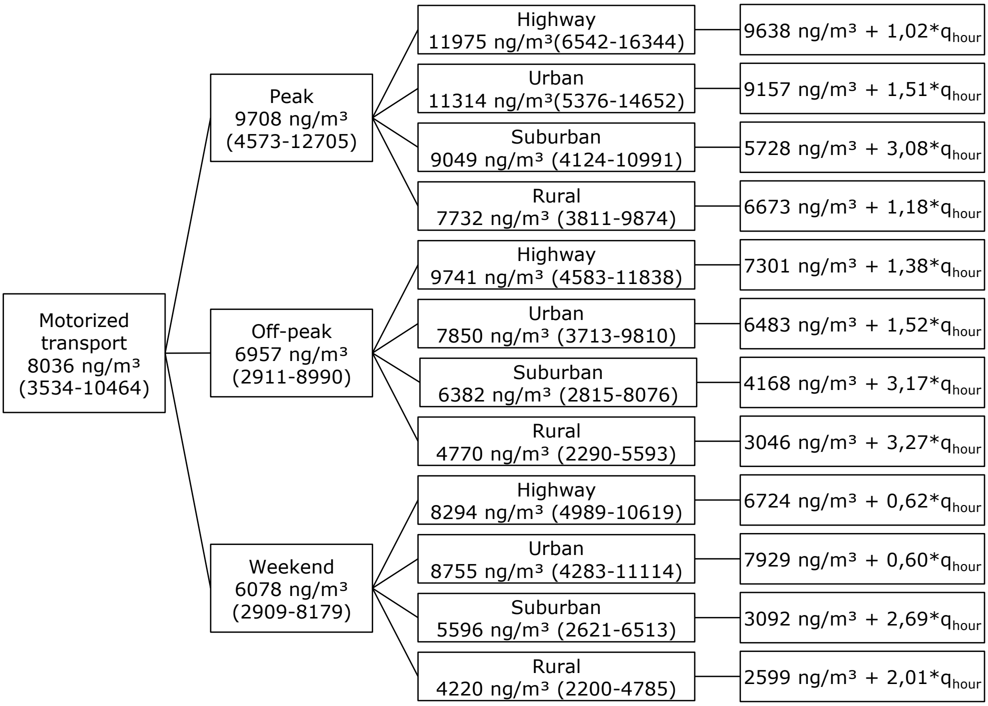


FIGURE A18: Visualization of the regression model for motorized transport (qhour is hourly traffic intensity). Peak hour is defined as 7-10 a.m. and 4-7 p.m. The tree should be used as a look-up table, e.g. if a car trip in peak hour in a rural area is considered, average exposure of the motorist is 7732 ng/m³. The interquartile range (ng/m³) is in brackets.

TABLE A9: Regression model for BC exposure for active travelers (R^2=0.02)

Variable	DF	Parameter Estimate	Standard Error	t Value	Pr > |t|
Intercept	1	2868	39.9	71.9	<.0001
Peak	1	1410	37.9	37.2	<.0001
Off-peak	1	830	37.5	22.2	<.0001
Urban	1	1029	37.7	27.3	<.0001
Suburban	1	405	37.4	10.9	<.0001

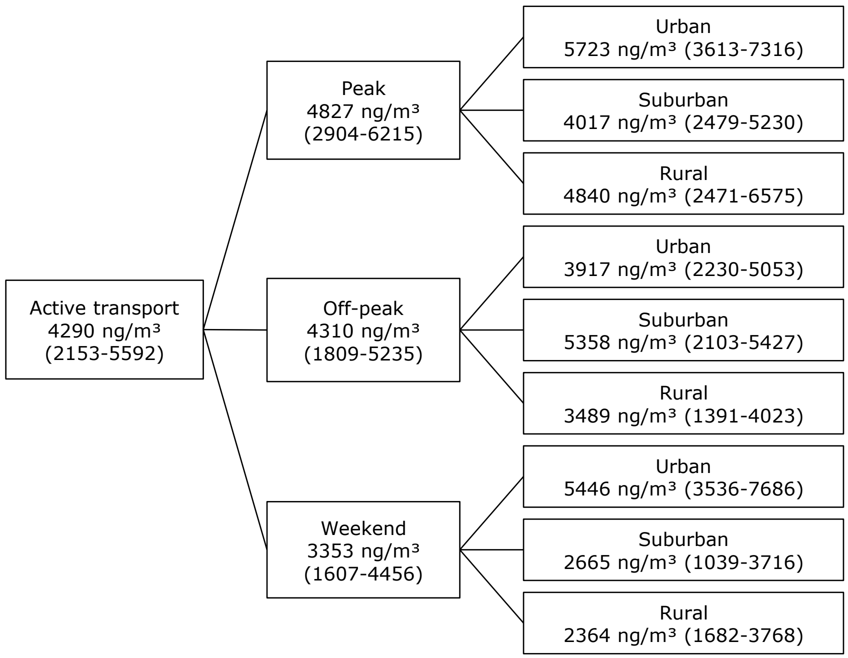


FIGURE A19: Visualization of the regression model for active transport. Peak hour is defined as 7-10 a.m. and 4-7 p.m. The tree should be used as a look-up table, e.g. if a bike trip in peak hour in a rural area is considered, average exposure of the cyclist is 4840 ng/m³. The interquartile range (ng/m³) is in brackets.

Sensitivity analysis – road type

One BC measurement is spread over maximum 300 GPS observations, and during one BC measurement streets with several road types are used. The dataset used in chapter 3.3 was adapted by considering whether at least 75% of the GPS-observations with the same BC measurement had the same road type (e.g. 5-min BC is 5000 ng/m³, if 15% of the observations is on a local road and 85% on a highway; only link the highway observations to the BC measurement). If less than 75% is on the same road type (e.g. 45% on highway and 55% on local road), the complete 5-min BC measurement was deleted.

As a consequence of the sensitivity analysis, 24% of 1-sec observations with motorized modes are deleted, whereas 9% of trips with active modes are deleted in the sensitivity. On highways, 18% of the motorized observations is deleted; 33% on major roads; and 22% on local roads. For active modes, 43% of the observations on major roads were deleted, and just 5% on local roads. FIGURE A20 and FIGURE A21 show the results with the reduced sensitivity dataset, and compare the results with the data presented in chapter 3.3. Although the substantial removal of observations due to the sensitivity analysis, the resulting average concentrations and boxplots are very similar to the results presented in the manuscript.

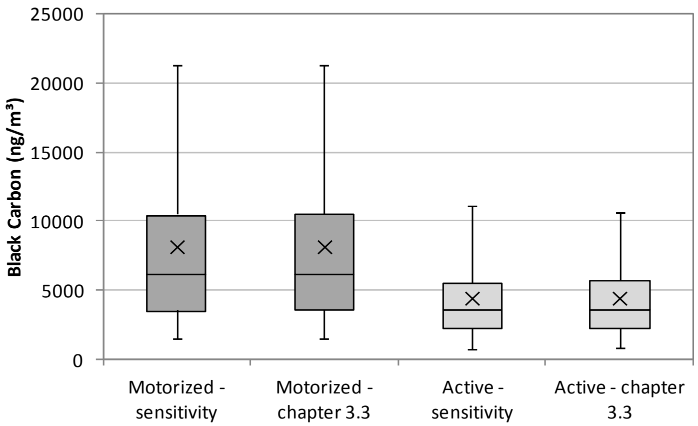


FIGURE A20: Results of the sensitivity analysis – impact on BC concentrations for motorists and active travelers. Boxplot showing 5th, 25th, 75th, 95th percentiles, median and mean (×).

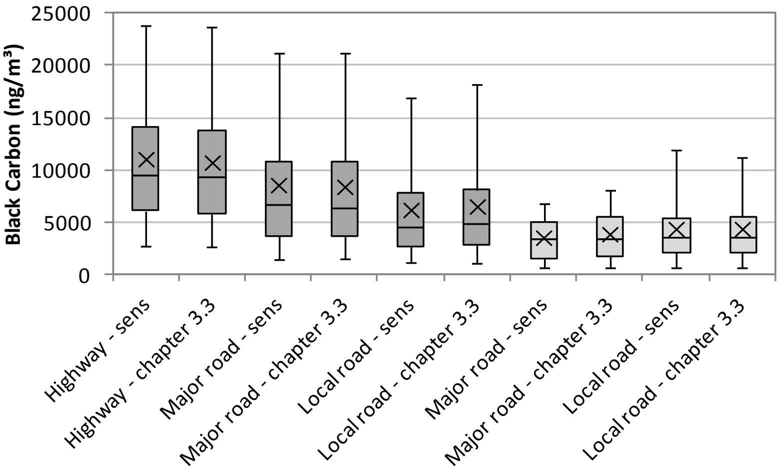


FIGURE A21: Results of the sensitivity analysis - BC exposure (ng/m³) is presented on the y-axis and this is related to transport mode and road type on the x-axis. Motorized modes (car, bus) are shown in darker gray, active modes (bike, on foot) in lighter gray. Represented are P5, 1st quartile, median, 3rd quartile and P95. The cross marks the mean value. 'Chapter 3.3' presents the results as they are included in chapter 3.3 of this book; 'sens' gives the results of the sensitivity analysis.

In summary, for active travelers, most trips use primarily the same road type (the activity space of an active traveler is limited in just 5 minutes); for motorized modes almost a quarter of all observations is cut in the sensitivity analysis meaning that for approximately one quarter of the observations road type changes within 5 minutes.

A.3 SUPPLEMENTAL MATERIAL CHAPTER 4.2

FIGURE A22: Aethalometer model AE51 (AethLabs, 2011) in a weatherproof housing. On most measurement sites an extra battery was placed inside the housing, so no other external power was necessary.

FIGURE A23: Measurement sites at building facades or on lamp posts.

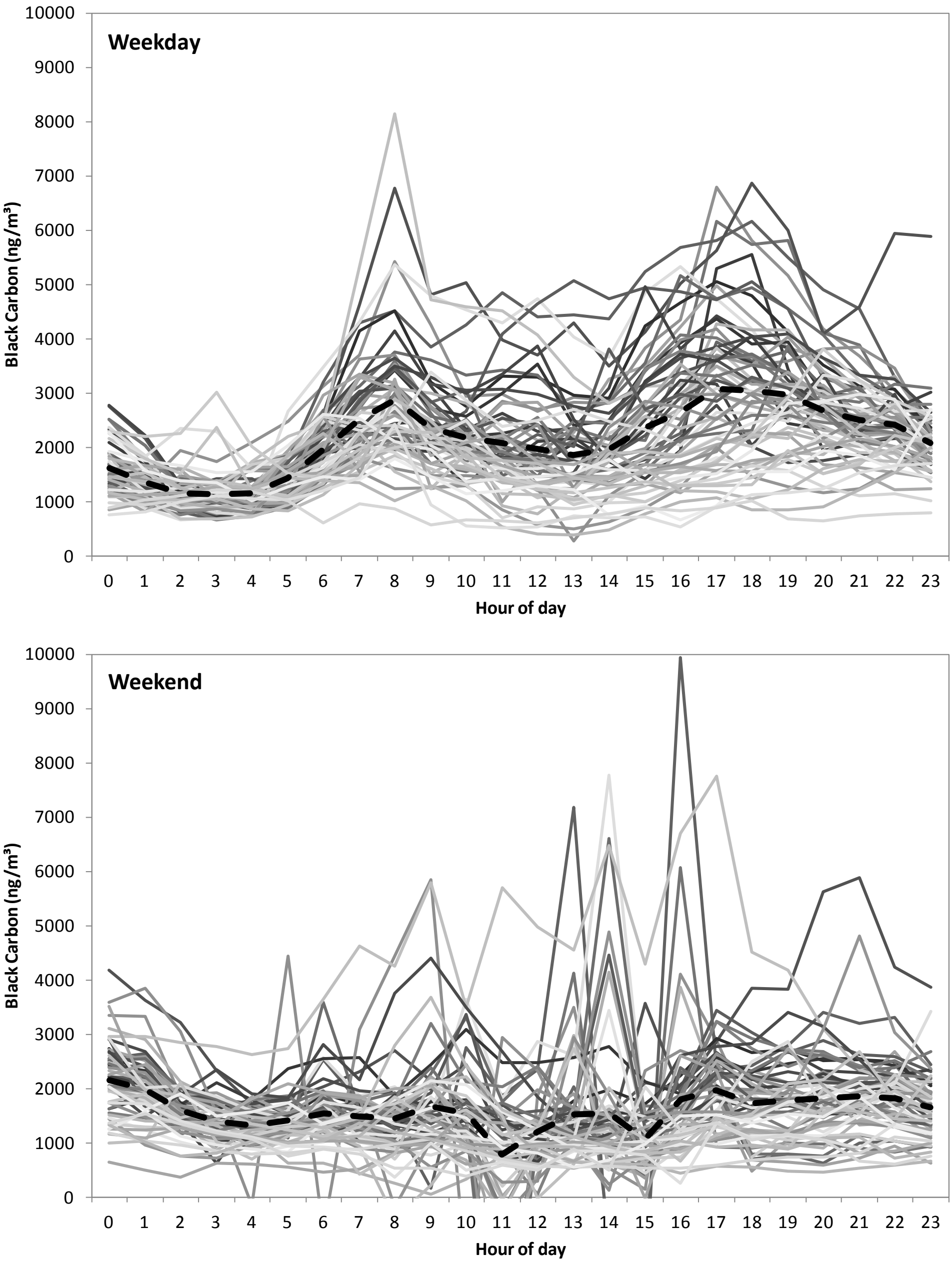


FIGURE A24: Hourly BC concentrations at 63 locations (upper: on 5 weekdays Monday to Friday; below: on 2 weekend days Saturday and Sunday). The dotted line shows the average of all locations.

TABLE A10: Results of BC sampling (ng/m³) averaged for weekday hours (WKDY) and weekend hours (WKND)

	Hour	N	Mean	Median	StdDev
All_hourly	0-23	3005	1861	1644	1037
WKDY	0	63	1608	1517	470
WKDY	1	63	1350	1263	357
WKDY	2	63	1150	1097	335
WKDY	3	63	1124	1034	398
WKDY	4	63	1153	1106	318
WKDY	5	63	1422	1433	400
WKDY	6	63	1963	1921	572
WKDY	7	63	2500	2273	860
WKDY	8	63	2875	2633	1235
WKDY	9	63	2357	2211	873
WKDY	10	63	2162	1991	856
WKDY	11	63	2069	1894	947
WKDY	12	63	1973	1740	1004
WKDY	13	63	1854	1685	955
WKDY	14	63	1967	1798	883
WKDY	15	63	2346	2115	1119
WKDY	16	63	2655	2515	1250
WKDY	17	63	3081	2975	1409
WKDY	18	63	3047	2989	1348
WKDY	19	63	2955	2917	1131
WKDY	20	63	2664	2724	852
WKDY	21	63	2495	2460	747
WKDY	22	63	2407	2371	720
WKDY	23	63	2077	2026	655
WKND	0	62	2158	2123	673
WKND	1	62	1986	1972	625
WKND	2	62	1586	1577	470
WKND	3	62	1381	1335	412
WKND	4	62	1308	1328	397
WKND	5	62	1402	1343	538
WKND	6	62	1529	1414	648
WKND	7	62	1477	1381	614
WKND	8	62	1438	1366	830
WKND	9	62	1669	1488	1066
WKND	10	62	1529	1411	863
WKND	11	62	780	929	1572
WKND	12	62	1206	1065	768
WKND	13	62	1525	1186	1098
WKND	14	62	1538	1104	1578
WKND	15	62	1065	979	822
WKND	16	62	1800	1454	1527
WKND	17	62	1961	1836	1001
WKND	18	63	1715	1582	762
WKND	19	63	1777	1754	757
WKND	20	63	1811	1636	821
WKND	21	63	1851	1719	845
WKND	22	63	1816	1811	651
WKND	23	62	1648	1621	600

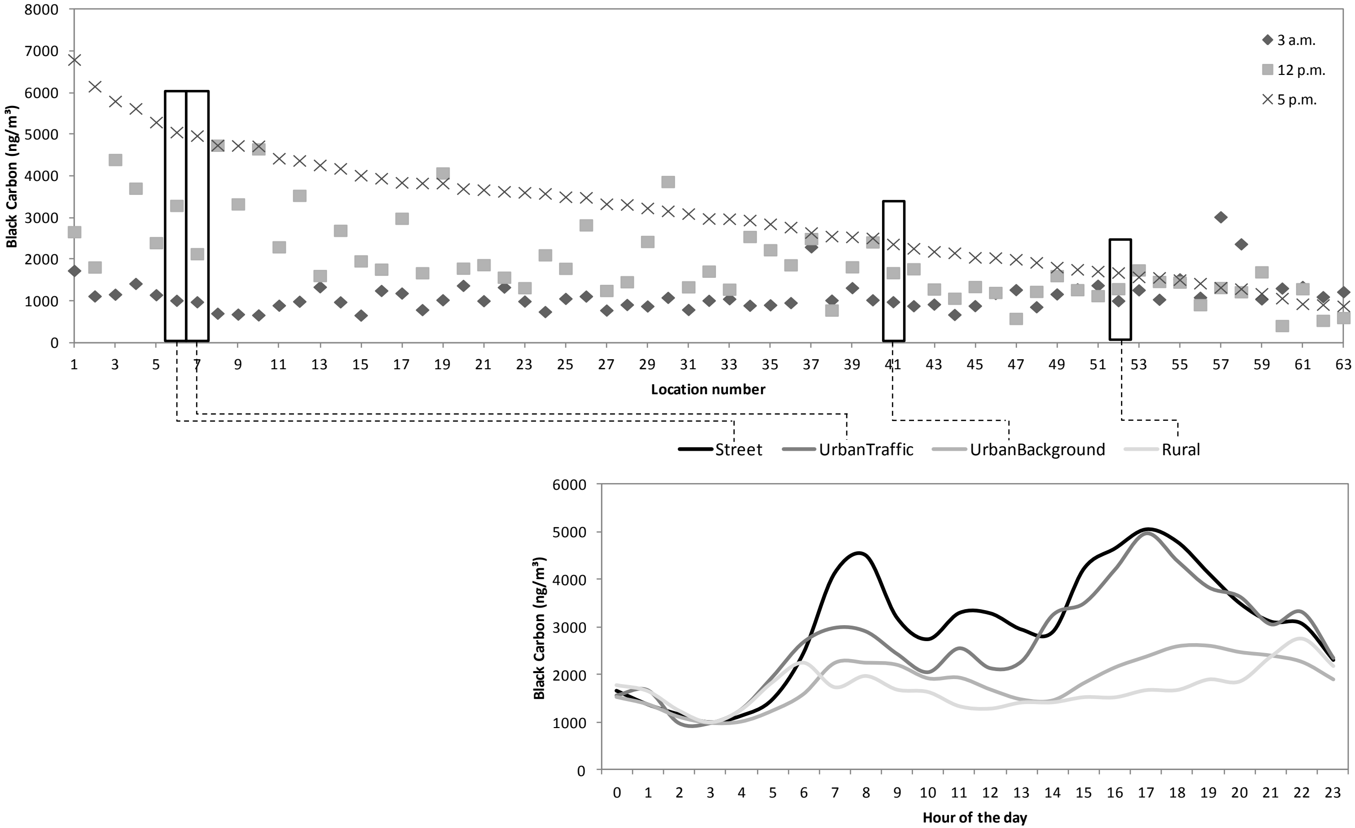


FIGURE A25: Top panel: Measured BC concentration on 63 locations for three hours (3 a.m., 12 p.m., 5 p.m., on weekdays), showing a different temporal pattern for different locations (ordered according to concentration at 5 p.m.). Below: Daily pattern in BC concentrations at 4 random locations, one of each type (Table SI1).

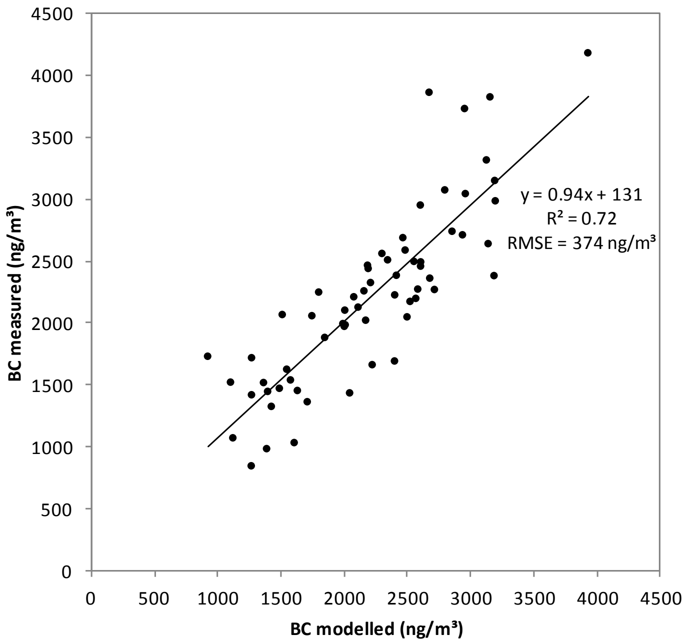


FIGURE A26: Leave-one-out cross-validation for the annual LUR model for BC

TABLE A11: Hourly LUR models for BC while keeping the significant variables of the annual LUR model (weekend hours) – HM1

						Highest Cook's D		ROADLENGTH_1000 [a]		TRAFLOADHV_FRACTION_100 [a]		TRAFLOAD_50_HEAVY [a]		DIST_NEAR [a]	
Day	hour	N	R^2	Adj R^2	RMSE		β_0	β_1	prob1	β_2	prob2	β_3	prob3	β_4	prob4
WKND	0	62	0.52	0.48	484.71	<1	917	0.03	0.00	-1777.63	0.31	300.23	0.06	-4.38	0.31
WKND	1	62	0.49	0.46	460.49	<1	911	0.03	0.00	-1242.88	0.46	323.99	0.03	-5.52	0.18
WKND	2	62	0.21	0.16	431.74	<1	1078	0.01	0.00	-365.27	0.82	160.03	0.25	-3.46	0.37
WKND	3	62	0.18	0.12	385.91	<1	1199	0.01	0.05	-1577.19	0.26	141.06	0.25	-7.66	0.03
WKND	4	62	0.08	0.01	393.93	<1	1055	0.01	0.05	-714.20	0.62	31.10	0.80	-1.97	0.57
WKND	5	62	0.17	0.11	506.28	<1	924	0.01	0.02	706.64	0.70	217.26	0.18	-3.47	0.44
WKND	6	62	0.17	0.11	609.48	<1	1037	0.01	0.07	3052.26	0.17	234.03	0.23	-2.80	0.60
WKND	7	62	0.25	0.20	549.41	<1	1019	0.01	0.04	1397.22	0.48	383.88	0.03	-7.15	0.15
WKND	8	62	0.17	0.11	783.46	<1	1442	0.00	0.83	1895.12	0.50	525.51	0.04	-9.56	0.17
WKND	9	62	0.11	0.05	1040.38	2.45	1613	0.00	0.99	1721.79	0.65	561.25	0.09	-9.84	0.29
WKND	10	62	0.17	0.12	811.72	<1	1315	0.00	0.63	6654.74	0.03	165.55	0.52	-7.24	0.32
WKND	11	62	0.07	0.01	1567.40	2.16	776	0.00	0.88	9918.89	0.08	-32.47	0.95	-6.39	0.65
WKND	12	62	0.37	0.33	628.46	<1	504	0.01	0.01	6294.26	0.01	299.14	0.14	-8.71	0.12
WKND	13	62	0.23	0.17	998.12	1.24	625	0.02	0.02	-1184.56	0.74	685.42	0.03	-12.84	0.15
WKND	14	62	0.38	0.34	1287.09	<1	633	0.02	0.13	25248.22	0.00	-751.38	0.07	-4.97	0.66
WKND	15	62	0.05	-0.01	826.65	<1	808	0.01	0.28	3281.45	0.28	-195.96	0.46	-5.05	0.49
WKND	16	62	0.29	0.24	1327.97	<1	446	0.03	0.01	-8441.15	0.08	1461.51	0.00	-14.43	0.22
WKND	17	62	0.32	0.27	853.09	1.42	711	0.03	0.00	-569.71	0.85	515.97	0.06	-10.42	0.17
WKND	18	63	0.34	0.29	641.50	1.05	685	0.02	0.00	1408.07	0.54	246.55	0.23	-7.01	0.22
WKND	19	63	0.28	0.23	663.36	<1	879	0.02	0.00	-687.33	0.77	314.43	0.14	-7.43	0.21
WKND	20	63	0.19	0.14	763.15	<1	1078	0.02	0.00	-2132.67	0.44	198.50	0.41	-8.84	0.19
WKND	21	63	0.16	0.10	802.66	<1	1168	0.02	0.01	-1103.25	0.70	45.50	0.86	-8.57	0.23
WKND	22	63	0.35	0.30	544.52	<1	869	0.02	0.00	403.40	0.84	87.70	0.61	-5.31	0.27
WKND	23	62	0.24	0.19	540.12	<1	833	0.02	0.00	1430.04	0.47	-28.84	0.87	-1.48	0.76
Year		63	0.77	0.75	356.84	2.58	812	0.03	0.00	4569.04	0.00	393.99	0.00	-8.06	0.01

[a] Total road length in a buffer with radius 1km (ROADLENGTH_1000), Fraction of traffic load that is heavy traffic in a buffer with radius 100m (TRAFLOADHV_FRACTION_100), Traffic load of heavy traffic in a buffer with radius 50m – indicator variable (TRAFLOAD_50_HEAVY), Distance to the nearest road (DIST_NEAR)

TABLE A12: Hourly LUR models for BC using hourly monitoring data and static independent variables: independent models (weekend hours) – HM2

hour	N	R^2	Adj R^2	RMSE	Highest Cook's D	β_0	β_1	X_1 [a]	β_2	X_2 [a]	β_3	X_3 [a]	β_4	X_4 [a]
0	62	0.47	0.46	493.43	<1	866	0.03054	ROADLENGTH_1000						
1	62	0.48	0.46	458.16	<1	655	0.02603	ROADLENGTH_1000	0.01154	Q_NEAR_MAJOR				
2	62	0.26	0.23	412.56	<1	1290	0.000314	HDRES_1000	0.00895	Q_NEAR_MAJOR				
3	62	0.21	0.18	373.41	<1	1418	0.000207	HDRES_1000	-7.8744	DIST_NEAR				
4	62	0.15	0.12	371.52	<1	1084	0.1276	ADDRESS_500	2.02557	QD_NEAR1_HEAVY				
5	62	0.27	0.23	471.14	1.19	773	4.8824	ADDRESS_100	302.664	TRAFLOAD_50_HEAVY	0.00985	Q_NEAR_MAJOR		
6	62	0.24	0.22	572.69	<1	1078	0.00234	TRAFLOAD_100_HEAVY	0.23052	ADDRESS_500				
7	62	0.45	0.41	472.84	1.17	53	425.626	TRAFLOAD_50_HEAVY	0.02802	ROADLENGTH_1000	-0.00019	URBGR_5000	0.000034	LDRES_5000
8	62	0.33	0.29	698.59	<1	422	622.402	TRAFLOAD_50_HEAVY	-0.00046	URBGR_3000	0.000053	LDRES_5000		
9	62	0.09	0.07	1027.03	<1	1461	678.384	TRAFLOAD_50_HEAVY						
10	62	0.17	0.15	794.66	<1	1387	0.03755	TRAFLOADHV_FRACTION_100_2						
11	62	0.24	0.20	1407.21	<1	1082	0.00544	TRAFLOAD_100_HEAVY	-0.00052	URBGR_3000	821.126	D_HIGH_LT1000		
12	62	0.48	0.44	573.04	1.74	30	8750.51	TRAFLOADHV_FRACTION_100	0.03489	ROADLENGTH_1000	-0.000119	URBGR_5000	0.000198	IND_3000
13	62	0.27	0.24	955.16	3.09	710	0.1388	ADDRESS_1000	13767	D_NEAR_MAJOR1				
14	62	0.37	0.34	1278.52	<1	84	19005	TRAFLOADHV_FRACTION_100	0.000083	LDRES_3000				
15	62	0.07	0.05	800.28	1.90	882	0.000245	IND_3000						
16	62	0.28	0.26	1313.61	1.08	501	0.69444	ADDRESS_500	1135.313	TRAFLOAD_50_HEAVY				
17	62	0.44	0.41	770.58	4.51	325	0.02953	ROADLENGTH_1000	7.44015	QD_NEAR1_HEAVY	0.00027587	IND_3000		
18	63	0.44	0.41	583.61	<1	592	0.03209	ROADLENGTH_1000	5.52691	QD_NEAR1_HEAVY	-0.000188	URBGR_3000		
19	63	0.37	0.35	611.38	<1	1425	6.45272	QD_NEAR1_HEAVY	0.000535	HDRES_1000				
20	63	0.32	0.29	689.67	1.46	1468	0.00063	HDRES_1000	4.54259	QD_NEAR1_HEAVY				
21	63	0.23	0.22	746.26	2.22	1624	0.00062	HDRES_1000						
22	63	0.39	0.37	515.53	<1	1257	0.04277	PEOPLE_1000	6190.905	D_NEAR_MAJOR1				
23	62	0.34	0.32	494.30	<1	1240	0.02568	PEOPLE_1000	0.00029	HDRES_1000				
Year	63	0.77	0.75	356.84	2.58	812	0.03	ROADLENGTH_1000	4569	TRAFLOADHV_FRACTIO N_100	394	TRAFLOAD_50_H EAVY	-8.06	DIST_NEAR

[a] Total road length in a buffer with size XXm (ROADLENGTH_XX), Traffic intensity (private cars) on the nearest major road (Q_NEAR_MAJOR), High density residential in a buffer with size XXm (HDRES_XX), Distance to the nearest road (DIST_NEAR), Number of addresses in a buffer with size XXm (ADDRESS_XX), Traffic intensity (heavy traffic) on the nearest road / distance to the nearest road (QD_NEAR1_HEAVY), Sum of (traffic intensity (heavy traffic) * road length) in a buffer with size XXm (TRAFLOAD_XX_HEAVY), Urban green in a buffer with size XXm (URBGR_XX), Low density residential in a buffer with size XXm (LDRES_XX), Fraction of heavy traffic squared in a buffer with size XXm (TRAFLOADHV_FRACTION_XX_2), Distance to the nearest highway < 1000m (D_HIGH_LT1000), Fraction of heavy traffic in a buffer with size XXm (TRAFLOADHV_FRACTION_XX), Industrial or commercial units in a buffer with size XXm (IND_XX), 1 / Distance to the nearest major road (D_NEAR_MAJOR1), Number of people in a buffer with size XXm (PEOPLE_XX)

TABLE A13: Hourly LUR models for BC using hourly monitoring data and dynamic independent variables: independent models (weekend hours) – HM3

hour	N	R^2	Adj R^2	RMSE	Highest Cook's D	β_0	β_1	X_1 [a]	β_2	X_2 [a]	β_3	X_3 [a]	β_4	X_4 [a]
0	62	0.47	0.46	493.43	<1	866	0.03054	ROADLENGTH_1000						
1	62	0.48	0.47	456.06	<1	707	0.02564	ROADLENGTH_1000	1.85097	**Q_NEAR_MAJOR_WKND1**				
2	62	0.31	0.29	397.49	<1	1214	0.000278	HDRES_1000	2.91379	**Q_NEAR_MAJOR_WKND2**				
3	62	0.21	0.18	373.41	<1	1418	0.000207	HDRES_1000	-7.8744	DIST_NEAR				
4	62	0.15	0.12	371.52	<1	1084	0.1276	ADDRESS_500	2.02557	QD_NEAR1_HEAVY				
5	62	0.23	0.20	480.32	2.59	1020	4.4095	ADDRESS_100	0.00084	**TRAFLOAD300_WKND5**				
6	62	0.24	0.22	572.69	<1	1078	0.00234	TRAFLOAD_100_HEAVY	0.23052	ADDRESS_500				
7	62	0.45	0.41	472.84	1.17	53	425.626	TRAFLOAD_50_HEAVY	0.02802	ROADLENGTH_1000	-0.00019	URBGR_5000	0.000034	LDRES_5000
8	62	0.33	0.29	698.59	<1	422	622.402	TRAFLOAD_50_HEAVY	-0.00046	URBGR_3000	0.000053	LDRES_5000		
9	62	0.09	0.07	1027.03	<1	1461	678.384	TRAFLOAD_50_HEAVY						
10	62	0.17	0.15	794.66	<1	1387	0.03755	TRAFLOADHV_FRACTION_100_2						
11	62	0.24	0.20	1407.21	<1	1082	0.00544	TRAFLOAD_100_HEAVY	-0.00052	URBGR_3000	821.126	D_HIGH_LT1000		
12	62	0.48	0.44	573.04	1.74	30	8750.51	TRAFLOADHV_FRACTION_100	0.03489	ROADLENGTH_1000	-0.000119	URBGR_5000	0.000198	IND_3000
13	62	0.27	0.24	955.16	3.09	710	0.1388	ADDRESS_1000	13767	D_NEAR_MAJOR1				
14	62	0.37	0.34	1278.52	<1	84	19005	TRAFLOADHV_FRACTION_100	0.000083	LDRES_3000				
15	62	0.07	0.05	800.28	1.90	882	0.000245	IND_3000						
16	62	0.28	0.26	1313.61	1.08	501	0.69444	ADDRESS_500	1135.313	TRAFLOAD_50_HEAVY				
17	62	0.44	0.41	770.58	4.51	325	0.02953	ROADLENGTH_1000	7.44015	QD_NEAR1_HEAVY	0.00027587	IND_3000		
18	63	0.44	0.41	583.61	<1	592	0.03209	ROADLENGTH_1000	5.52691	QD_NEAR1_HEAVY	-0.000188	URBGR_3000		
19	63	0.37	0.35	611.38	<1	1425	6.45272	QD_NEAR1_HEAVY	0.000535	HDRES_1000				
20	63	0.32	0.29	689.67	1.46	1468	0.00063	HDRES_1000	4.54259	QD_NEAR1_HEAVY				
21	63	0.23	0.22	746.26	2.22	1624	0.00062	HDRES_1000						
22	63	0.38	0.36	520.03	<1	1266	0.04160	**PEOPLE_1000_WKND22**	6377.289	D_NEAR_MAJOR1				
23	62	0.36	0.34	487.89	<1	893	0.00074	HDRES_1000	0.00092	LDRES_500				
Year	63	0.77	0.75	356.84	2.58	812	0.03	ROADLENGTH_1000	4569	TRAFLOADHV_FRACTION_100	394	TRAFLOAD_50_HEAVY	-8.06	DIST_NEAR

Dynamic variables that enter the model are indicated in bold.

[a] Total road length in a buffer with size XXm (ROADLENGTH_XX), Traffic intensity (private cars) on the nearest major road at hour x (Q_NEAR_MAJOR_WKNDx), High density residential in a buffer with size XXm (HDRES_XX), Distance to the nearest road (DIST_NEAR), Number of addresses in a buffer with size XXm (ADDRESS_XX), Traffic intensity (heavy traffic) on the nearest road / distance to the nearest road (QD_NEAR1_HEAVY), Sum of (traffic intensity (private cars) at hour x * road length) in a buffer with size XXm (TRAFLOADXX_WKNDx), Sum of (traffic intensity (heavy traffic) * road length) in a buffer with size XXm (TRAFLOAD_XX_HEAVY), Urban green in a buffer with size XXm (URBGR_XX), Low density residential in a buffer with size XXm (LDRES_XX), Fraction of heavy traffic squared in a buffer with size XXm (TRAFLOADHV_FRACTION_XX_2), Distance to the nearest highway < 1000m (D_HIGH_LT1000), Fraction of heavy traffic in a buffer with size XXm (TRAFLOADHV_FRACTION_XX), Industrial or commercial units in a buffer with size XXm (IND_XX), 1 / Distance to the nearest major road (D_NEAR_MAJOR1), Number of people in a buffer with size XXm (PEOPLE_XX)

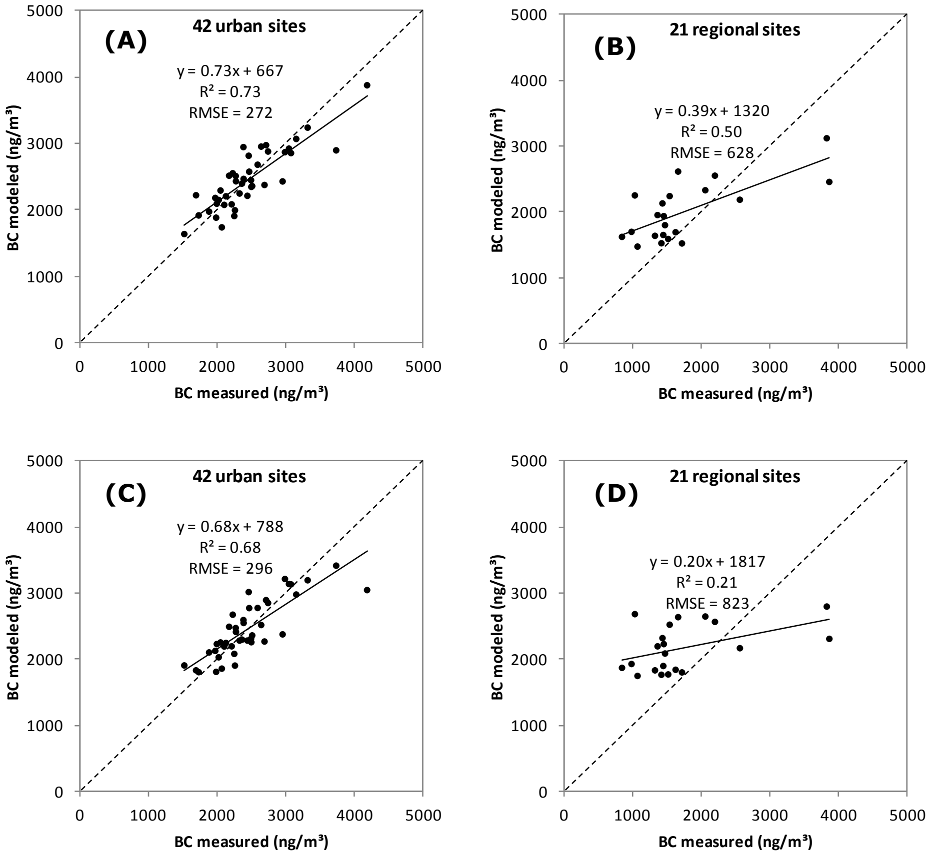


FIGURE A27: Annual model with high Cook's D values (A, B) and annual model with Cook's D values < 1 (C, D). The models are estimated with 42 urban points (A, C) and validated with 21 points of the regional monitoring campaign (B, D).

A.4 SUPPLEMENTAL MATERIAL CHAPTER 4.3

Example diaries: output from the activity-based model FEATHERS

The activity-based model produces diaries for every agent in a population, including the geographical subzone where each activity is done. For the moment FEATHERS produces diaries for the adult population, a population of approximately 6 million people. Four example diaries are presented in TABLE A14.

TABLE A14: Four example diaries as produced by the FEATHERS activity-based model. For every 'Person' (column PersonCounter) additional information is provided: subzone of residence, gender, person age, driver's license, worker or homemaker, age of oldest member of the household, number of cars in the household, socio-economic class, household composition. Trips are defined together with the activity performed at the destination side. The model predicts a diary starting from 3 a.m. (Beginning Time: 300) up to the next day 3 a.m.
Person 1 starts his day with a home-based activity at 3 a.m.; he makes a trip with his bike or on foot at 7.55 a.m. with a total duration of 5 minutes. He is in a shop from 8 a.m. for 70 minutes, and returns home again for 5 minutes with active modes. He spends the rest of the day at home.

Household Counter	Person Counter	Day [a]	Activity Type [b]	Beginning Time	Duration (min)	Location (subzone)	Trip Duration (min)	Transport Mode [c]
1	1	1	0	300	295	110	0	999999
1	1	1	4	800	70	110	5	3
1	1	1	0	915	1065	110	5	3
1	2	1	0	300	103	110	0	999999
1	2	1	1	624	495	2307	101	4
1	2	1	4	1627	75	108	108	4
1	2	1	4	1747	144	108	5	4
1	2	1	0	2152	308	110	101	4
2	3	1	0	300	797	110	0	999999
2	3	1	3	1622	5	110	5	1
2	3	1	0	1632	74	110	5	1
2	3	1	8	1749	49	109	3	1
2	3	1	0	1841	499	110	3	1
2	4	1	0	300	238	110	0	999999
2	4	1	1	702	542	111	4	1
2	4	1	4	1608	59	99	4	1
2	4	1	8	1712	103	99	5	1
2	4	1	4	1900	30	99	5	1
2	4	1	0	1936	444	110	6	1

[a] 1 Monday; 2 Tuesday; 3 Wednesday; 4 Thursday; 5 Friday; 6 Saturday; 7 Sunday
[b] 0 Home-based activity; 1 Work/education; 2 Business; 3 Bring/get; 4 Shopping; 6 Services; 7 Social; 8 Leisure; 9 Touring; 10 Other
[c] 1 Car driver; 3 Active modes; 4 Public transport; 6 Car passenger

Determination of concentration in subzones

- Hourly LUR models

Hourly models, independent of each other, can be developed by changing the dependent variable to reflect different hours of the day; but significant variables may vary from hour to hour. This results in hourly R^2 values of 0.07 – 0.8 and RMSE values of 287 ng/m³ – 1407 ng/m³. During the day traffic variables (heavy traffic and distance to the nearest (major) road) often enter the model. Traffic intensity (light traffic only) is significant on peak hours (8 a.m., 9 a.m., 5 p.m., 7 p.m.). On weekday nights, land use variables in larger buffers come into the model, but the limited concentration contrast results in low R^2, and the concentrations are mainly predicted by the intercept. On weekend nights BC concentrations were elevated and traffic variables are significant, indicative of busy traffic on weekend nights. Even though the hourly LUR models are developed independently of adjacent hours, in fact similar variables often return in consecutive models demonstrating the robustness of the models.
The hourly LUR models are discussed in more detail in chapter 4.2.

- Application of the hourly LUR models in the subzones

The study area is divided into 2386 subzones with an average size of 5.7 km². There are on average 1157 addresses/buildings (IQR: 324-1543; median 767) in each subzone. For each subzone, a concentration needs to be defined that can be used in the following steps of the AB²C model. To limit computer runtime, it was explored how many random addresses had to be selected, to determine the median concentration on an address in that subzone with enough reliability. For 11 subzones (both urban and rural), we predicted median annual concentrations based on 100 random address points in the subzone, and compared it to the prediction with only 10 random points. Differences of about 200 ng/m³ are possible, but overall with very good correspondence. A random selection of 10 addresses was decided to be sufficient to determine exposure in a whole subzone (FIGURE A28).

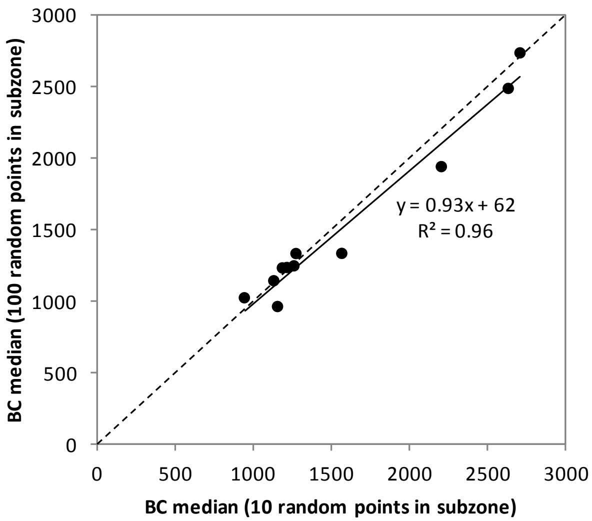


FIGURE A28: Median BC concentration in 11 subzones; once determined from 10 random addresses, and once from 100 random addresses.

BC concentrations were determined for approximately 23,860 addresses in the study area (FIGURE A29). There are a few subzones (11 zones) that do not have addresses in it; for these zones a BC concentration of 0 ng/m³ was assigned. It should be noted that FEATHERS-agents seldom perform activities in these zones. If 'Distance to nearest road' is a significant predictor in the hourly LUR model, this variable is truncated to 100m which is the maximum value in the calibration dataset, to prevent exposure estimates becoming negative. Where the regression equations produced negative estimates, values were set to 0 ng/m³ (most negative estimates occur on weekend days. E.g. at 11 a.m.: 4.08% of points were set to 0 ng/m³) (similar to (Henderson et al., 2007)). Where estimates exceeded the maximum measured concentrations by more than 20%, concentration estimates were truncated (on weekdays at 9 a.m., truncation was applied to 1.19% of the points) (similar to (Henderson et al., 2007)).

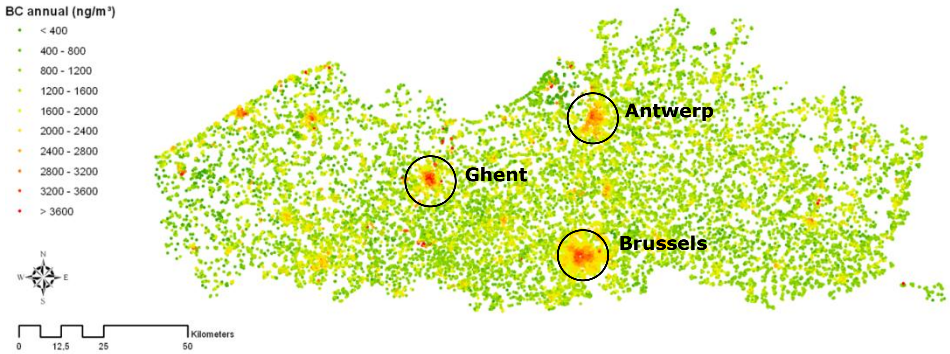


FIGURE A29: Predicted annual average BC concentrations for 10 random address points per subzone. The largest urban areas are indicated.

BC concentrations were determined for approximately 23,860 addresses in the study area, and the median concentration in each subzone was then defined as the exposure on fixed locations in that subzone (FIGURE A30).

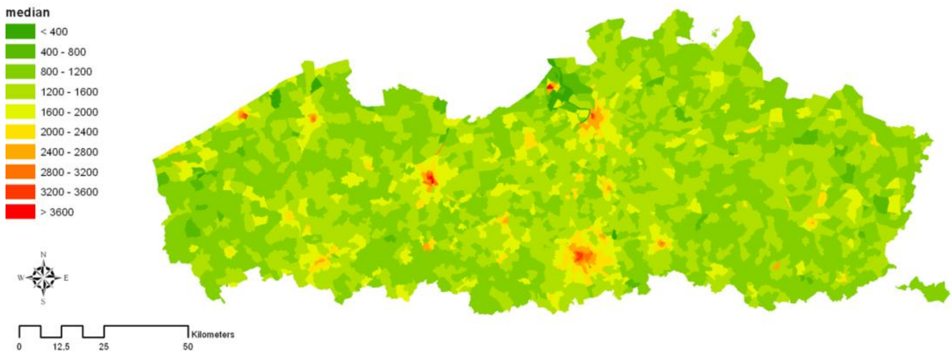
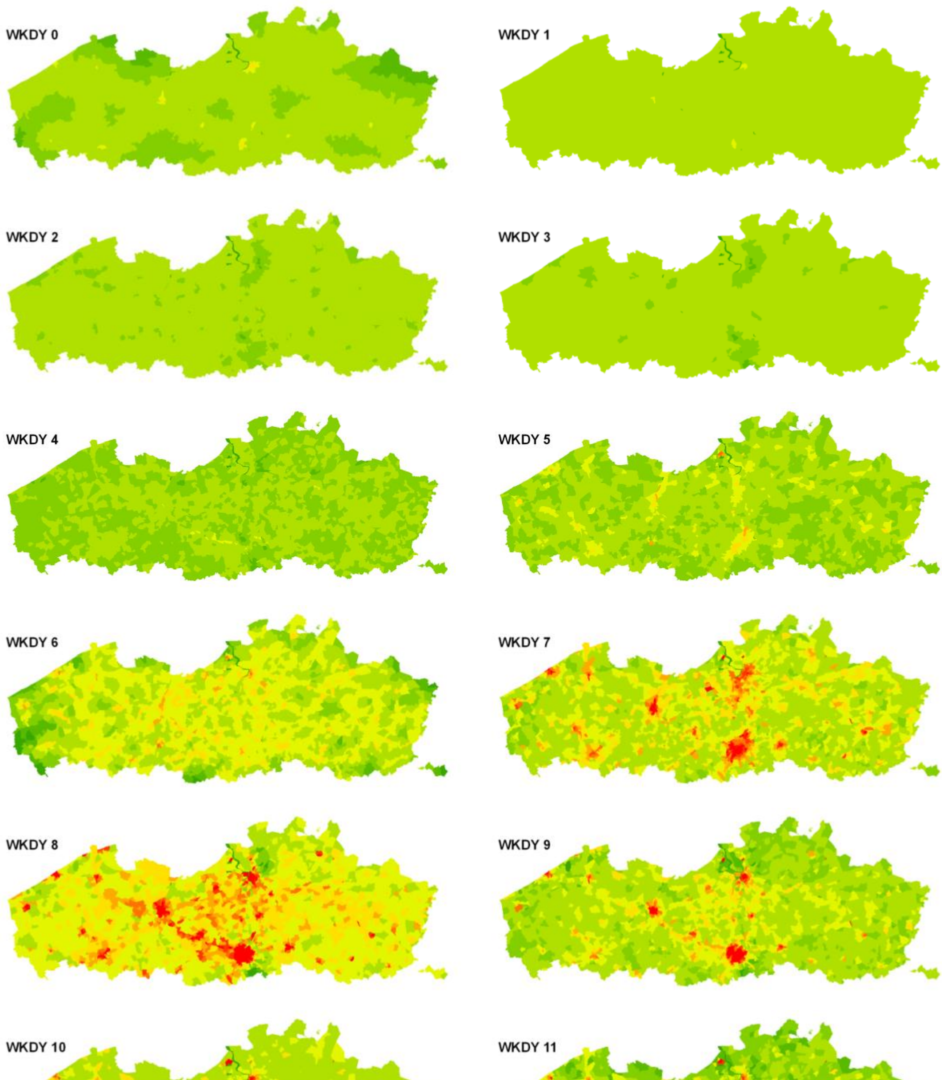
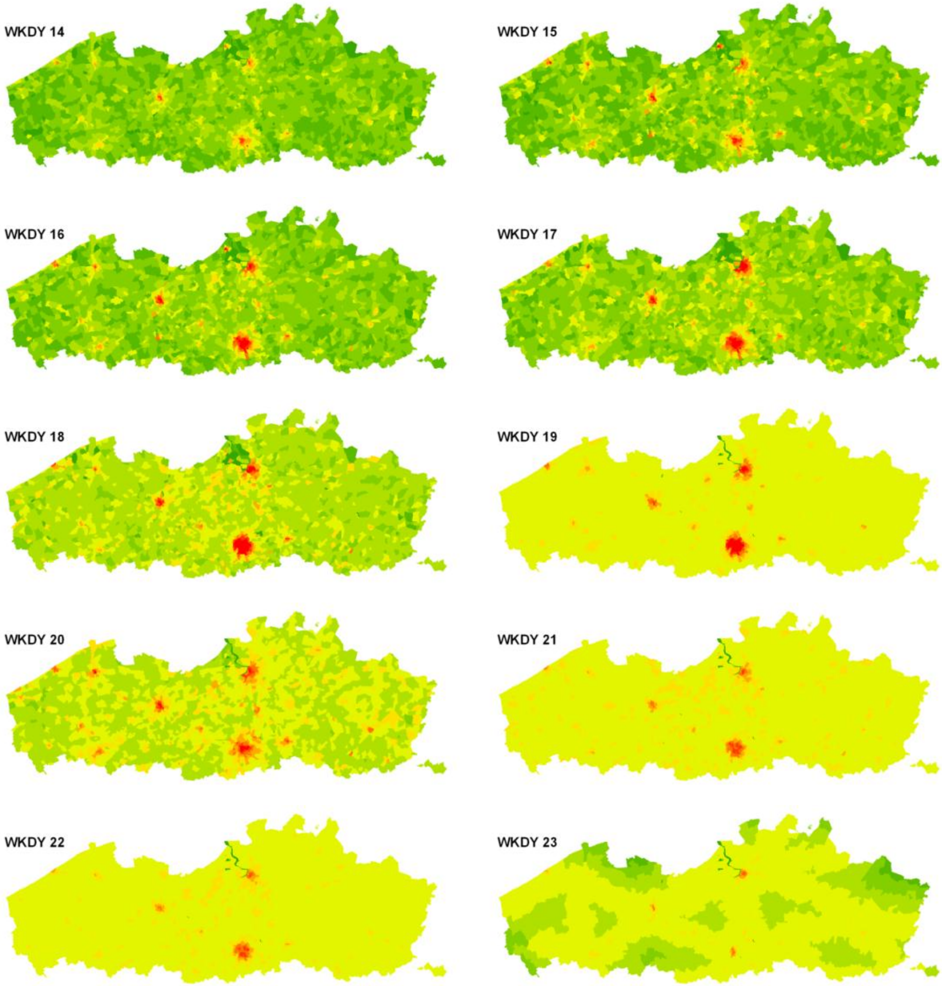
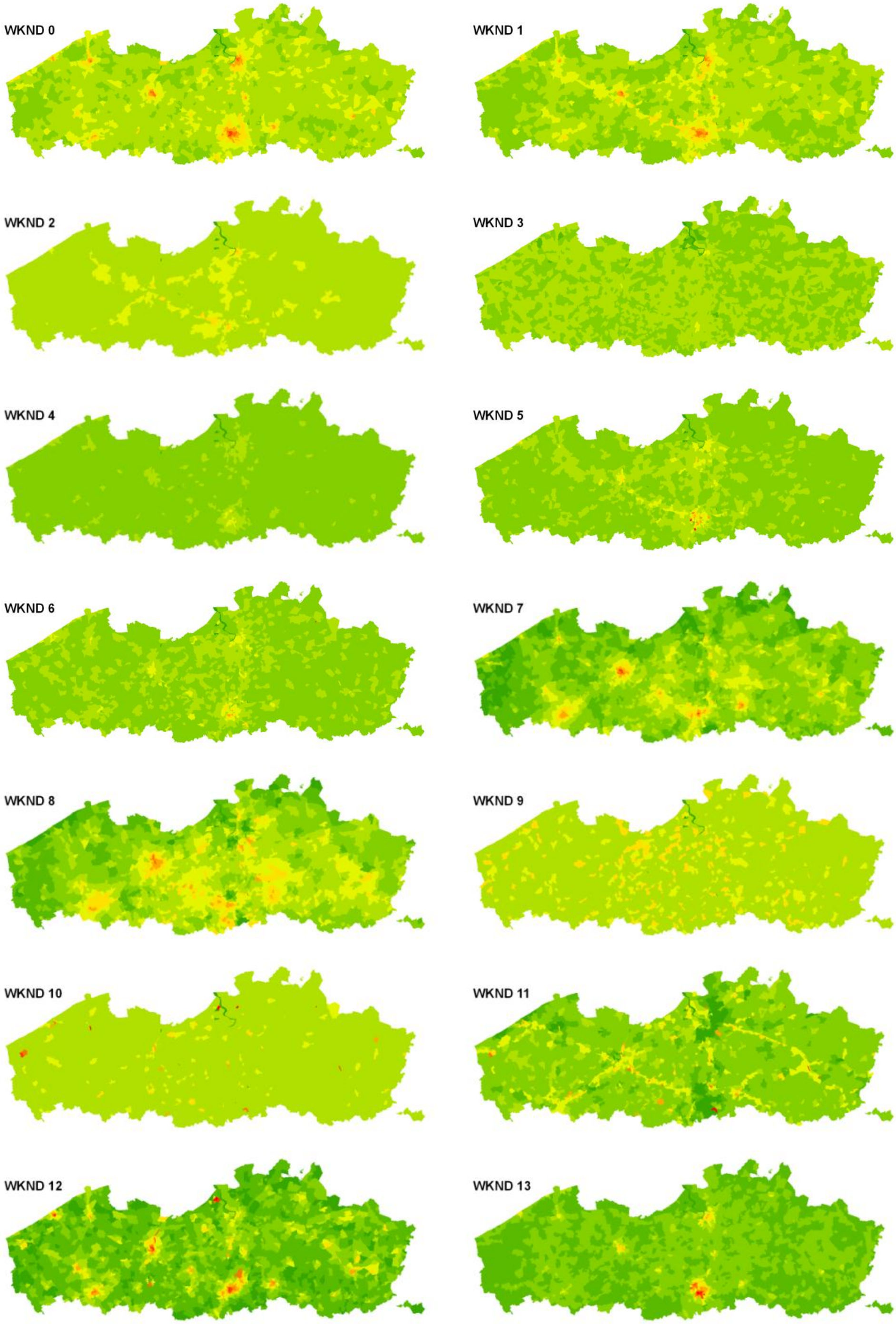


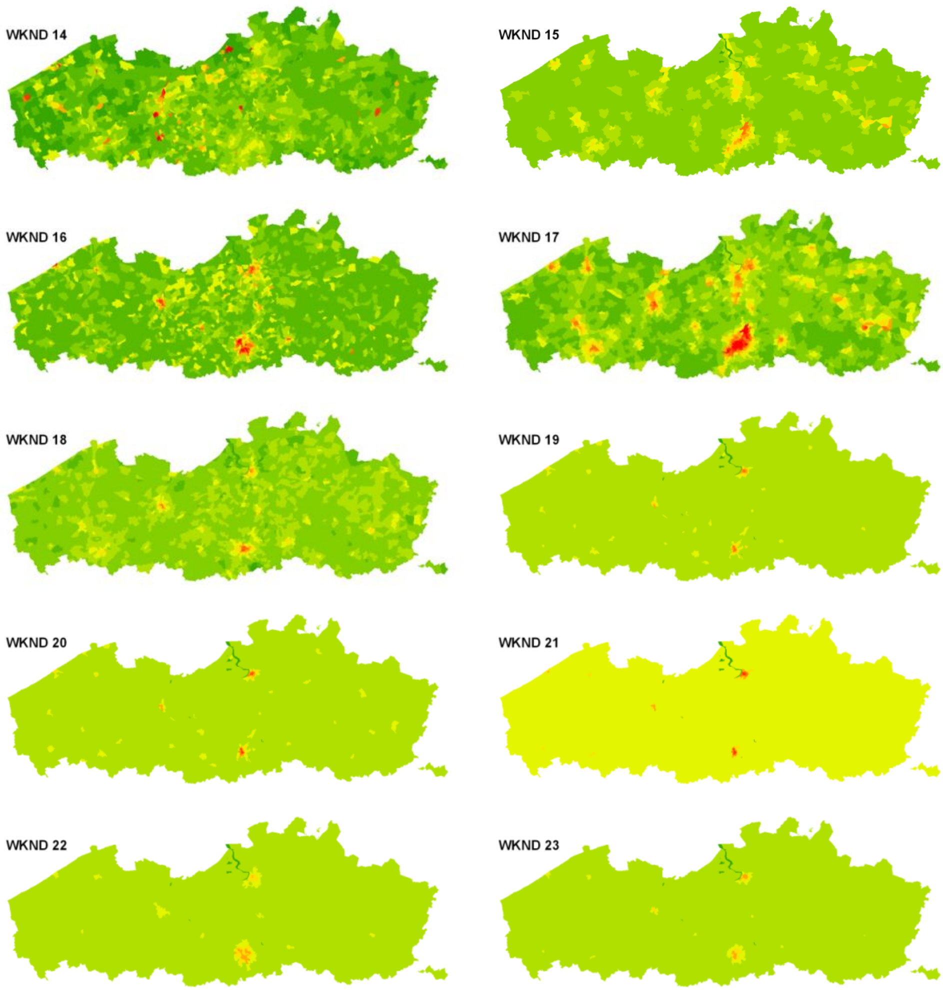
FIGURE A30: Median BC exposure (ng/m³) for every subzone defined in FEATHERS; calculated based on 10 randomly chosen addresses in each subzone. This is an *exposure* map as concentrations are defined on addresses and not on random points in each subzone (i.e. points are often situated near roads and not in the middle of a forest were concentrations are lower).

A similar approach as illustrated above with the annual average LUR-model, was followed for all hourly LUR models for weekdays and weekend days. The resulting hourly exposure maps are illustrated below (the same legend as in FIGURE A30 is applicable).


WKDY 0
WKDY 1
WKDY 2
WKDY 3
WKDY 4
WKDY 5
WKDY 6
WKDY 7
WKDY 8
WKDY 9
WKDY 10
WKDY 11
WKDY 12
WKDY 13

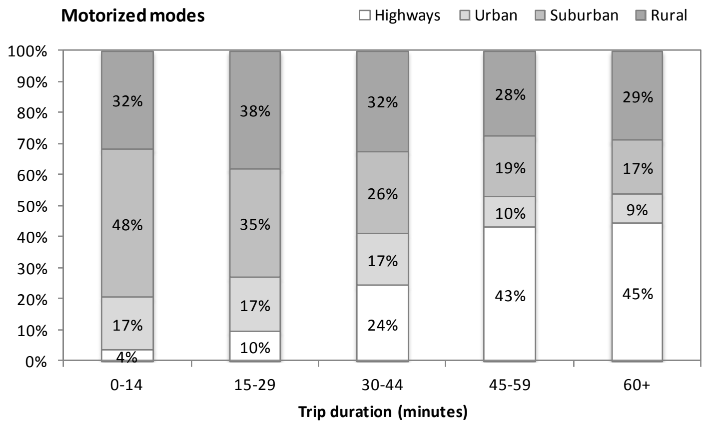

WKDY 14
WKDY 15
WKDY 16
WKDY 17
WKDY 18
WKDY 19
WKDY 20
WKDY 21
WKDY 22
WKDY 23

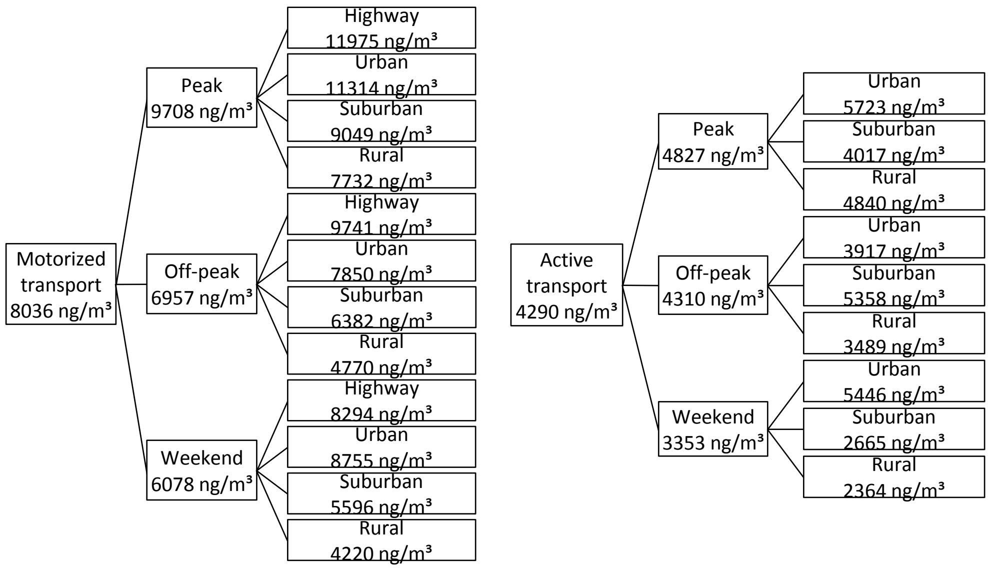

WKND 0
WKND 1
WKND 2
WKND 3
WKND 4
WKND 5
WKND 6
WKND 7
WKND 8
WKND 9
WKND 10
WKND 11
WKND 12
WKND 13


WKND 14
WKND 15
WKND 16
WKND 17
WKND 18
WKND 19
WKND 20
WKND 21
WKND 22
WKND 23

Concentration based on (chapter 3.3 and appendix A.2):

- Transport mode: motorized modes; active modes; public transport
- Timing: peak (weekdays: 7-8 a.m.; 8-9 a.m.; 9-10 a.m.; 4-5 p.m.; 5-6 p.m.; 6-7 p.m.); off-peak; weekend
- Urbanization based on proxy ‘trip duration’ (motorized) or subzone of origin (active modes)





Motorized transport: based on chapter 3.3. Degree of urbanization is unknown, so trip duration is used as proxy.

- If (tm=motorized and time=**Peak** and **dur<15**) then BC = ((0.04*11975) + (0.17*11314) + (0.48*9049) + (0.32*7732)) = 9220 ng/m^3

  If (tm=motorized and time=**Peak** and (**dur>14 and dur<30**)) then BC = ((0.10*11975) + (0.17*11314) + (0.35*9049) + (0.38*7732)) = 9226 ng/m^3

  If (tm=motorized and time=**Peak** and (**dur>29 and dur<45**)) then BC = ((0.24*11975) + (0.17*11314) + (0.26*9049) + (0.32*7732)) = 9624 ng/m^3

  If (tm=motorized and time=**Peak** and (**dur>44 and dur<60**)) then BC = ((0.43*11975) + (0.10*11314) + (0.19*9049) + (0.28*7732)) = 10165 ng/m^3

  If (tm=motorized and time=**Peak** and **dur>59**) then BC = ((0.45*11975) + (0.09*11314) + (0.17*9049) + (0.29*7732)) = 10188 ng/m^3
- If (tm=motorized and time=**Off-peak** and **dur<15**) then BC = ((0.04*9741) + (0.17*7850) + (0.48*6382) + (0.32*4770)) = 6314 ng/m^3

  If (tm=motorized and time=**Off-peak** and (**dur>14 and dur<30**)) then BC = ((0.10*9741) + (0.17*7850) + (0.35*6382) + (0.38*4770)) = 6355 ng/m^3

  If (tm=motorized and time=**Off-peak** and (**dur>29 and dur<45**)) then BC = ((0.24*9741) + (0.17*7850) + (0.26*6382) + (0.32*4770)) = 6858 ng/m^3

  If (tm=motorized and time=**Off-peak** and (**dur>44 and dur<60**)) then BC = ((0.43*9741) + (0.10*7850) + (0.19*6382) + (0.28*4770)) = 7522 ng/m^3

  If (tm=motorized and time=**Off-peak** and **dur>59**) then BC = ((0.45*9741) + (0.09*7850) + (0.17*6382) + (0.29*4770)) = 7558 ng/m^3
- If (tm=motorized and time=**Weekend** and **dur<15**) then BC = ((0.04*8294) + (0.17*8755) + (0.48*5596) + (0.32*4220)) = 5857 ng/m^3

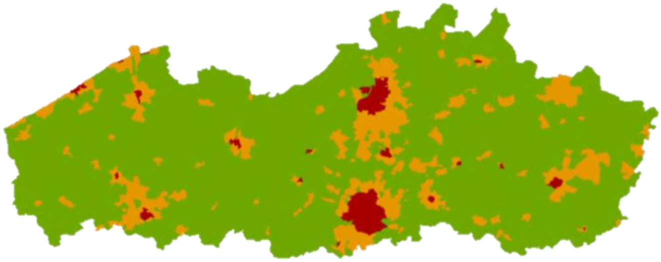
  If (tm=motorized and time=**Weekend** and (**dur>14 and dur<30**)) then BC = ((0.10*8294) + (0.17*8755) + (0.35*5596) + (0.38*4220)) = 5880 ng/m^3

  If (tm=motorized and time=**Weekend** and (**dur>29 and dur<45**)) then BC = ((0.24*8294) + (0.17*8755) + (0.26*5596) + (0.32*4220)) = 6284 ng/m^3

  If (tm=motorized and time=**Weekend** and (**dur>44 and dur<60**)) then BC = ((0.43*8294) + (0.10*8755) + (0.19*5596) + (0.28*4220)) = 6687 ng/m^3

  If (tm=motorized and time=**Weekend** and **dur>59**) then BC = ((0.45*8294) + (0.09*8755) + (0.17*5596) + (0.29*4220)) = 6695 ng/m^3

For **active modes**, the degree of urbanization of a trip will be determined by the degree of urbanization of the subzone of origin. Subzones were assigned a degree of urbanization (1=Urban; 2=Suburban; 3=Rural; no highway). Total road length in every category in a subzone is calculated, the category with the largest road length is assigned as degree of urbanization of that subzone. If a subzone has no roads, the centroid of the subzone is assigned to the nearest road and urbanization of that road is assigned to the subzone. As this map only covers Flanders, subzones in Brussels are automatically assigned urbanization=1 (see map).



- If (tm=active and time=**Peak** and urbanization=**Urban**) then BC = 5723 ng/m³
  If (tm=active and time=**Peak** and urbanization=**Suburban**) then BC = 4017 ng/m³
  If (tm=active and time=**Peak** and urbanization=**Rural**) then BC = 4840 ng/m³
- If (tm=active and time=**Off-peak** and urbanization=**Urban**) then BC =3917 ng/m³
  If (tm=active and time=**Off-peak** and urbanization=**Suburban**) then BC = 5358 ng/m³
  If (tm=active and time=**Off-peak** and urbanization=**Rural**) then BC = 3489 ng/m³
- If (tm=active and time=**Weekend** and urbanization=**Urban**) then BC = 5446 ng/m³
  If (tm=active and time=**Weekend** and urbanization=**Suburban**) then BC = 2665 ng/m³
  If (tm=active and time=**Weekend** and urbanization=**Rural**) then BC = 2364 ng/m³

For trips using **public transport** a fixed exposure of 3521 ng/m³ is assigned, derived from the personal measurement campaign. A model was not developed due to the limited number of observations in public transport.

Indoor air model

I/O-ratio: BC_Indoor = 0.76 * BC_Outdoor

The I/O-ratio is based on daily average I/O-ratios of individual homes (FIGURE A31). We did not use the original 5-min values to avoid strong local and temporary effects, and also because there is a delay before an increase outside is measured inside. In total we have 153 daily measurements in 24 different homes on which basis we determine the I/O-ratio. Homes with ETS were excluded from the study beforehand.

In the warm season the I/O-ratio is higher (0.84) compared to the cold season (0.74), which is in line with our expectations (more exchange of indoor and outdoor air because of increased ventilation).

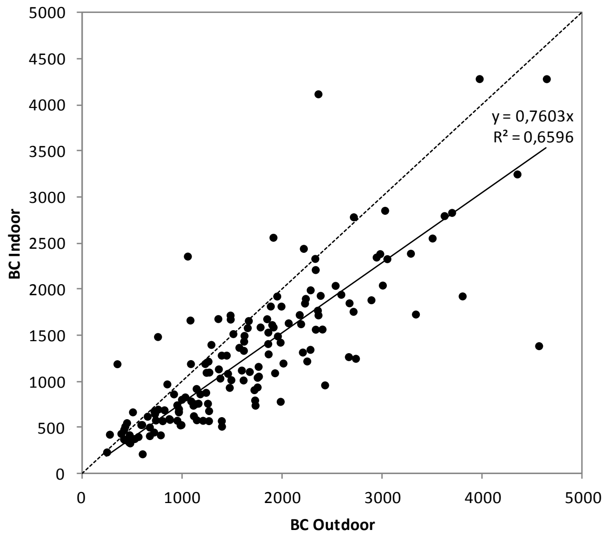


FIGURE A31: Indoor/Outdoor ratio

Validation dataset

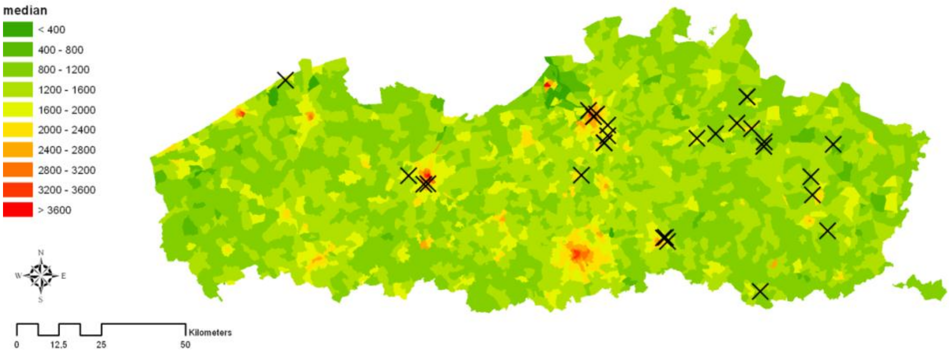


FIGURE A32: Median BC exposure (ng/m³) for every subzone defined in FEATHERS (FIGURE SI3) with indication of residential addresses of the 62 volunteers from the validation dataset

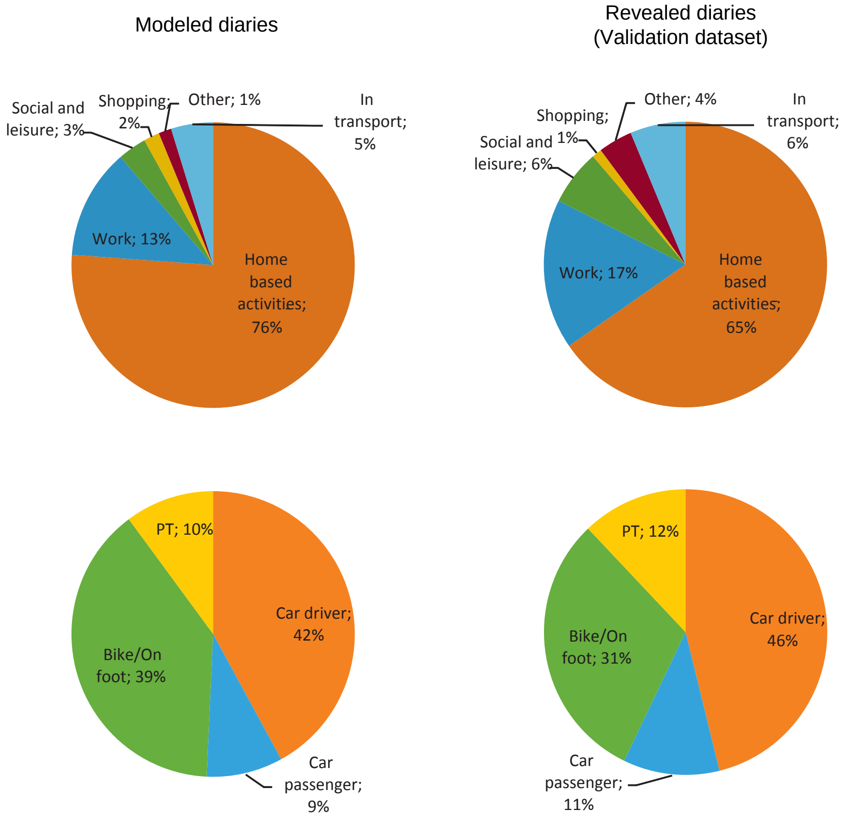


FIGURE A33: Comparison between modeled diaries (Left pie charts; 54 unique participants; based on 100 diaries per participant per day (Mon-Sun)) and revealed diaries (Right pie charts; 62 volunteers; based on 1 diary per participant per day (Mon-Sun)). PT=Public Transport (train, bus, light rail/metro).

Predicted and observed exposures: aggregated analysis

TABLE A15: Average exposure per activity

BC [ng/m³]	Observed	Predicted
Touring	2855	3822
Social and leisure	2445	1350
Shopping & services	2540	1339
Other	1829	1346
Being at home	1214	1211
Work	1077	1321
In transport	5132	6430

TABLE A16: Average exposure per transport mode

BC [ng/m³]	Observed	Predicted
Car driver	6432	7258
Car passenger	5583	7180
Active modes	3365	4124
Public transport	3521	3521

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PERSONAL INFORMATION

Place of birth:	Antwerp (Wilrijk), Belgium
Date of birth:	July 17, 1986
Nationality:	Belgian

EDUCATION

2007 – 2009	*Master of Transportation Sciences, specialization in Mobility Management* Hasselt University, Belgium
2004 – 2007	*Bachelor of Transportation Sciences* Hasselt University, Belgium

WORK EXPERIENCE

Nov 2009 – Nov 2013	Hasselt University (Transportation Research Institute) and VITO (Flemish Institute for Technological Research) PhD in Transportation Sciences: "Air pollution exposure assessment through personal monitoring and activity-based modeling" Supervisors: Prof. dr. G. Wets and Prof. dr. L. Int Panis
Jan 2010 – Sep 2012	Contributed to the HEAPS project (Health Effects of Air Pollution in Antwerp Schools). The study was financed by the Flemish Environment Agency (VMM), Flemish Administration of Environment, Nature and Energy (LNE), and Flemish Agency for Care and Health (VAZG).
Jun 2008 – Dec 2012	Contributed to the FP7-ESCAPE project (European Study of Cohorts for Air Pollution Effects).

JOURNAL PUBLICATIONS

Year	Journal	Information
2013	Environment International (IF 6.248)	Authors: *Sofie De Prins, Gudrun Koppen, Griet Jacobs, Evi Dons, Els Van de Mieroop, Vera Nelen, Frans Fierens, Luc Int Panis, Patrick De Boever, Bianca Cox, Tim Nawrot, Greet Schoeters* Title: *Influence of ambient air pollution on global DNA methylation in healthy adults: a seasonal follow-up* Volume, pages: *59, 418-424*
2013	Atmospheric Environment (IF 3.110)	Authors: *Wouter Lefebvre, Martine Van Poppel, Bino Maiheu, Stijn Janssen, Evi Dons* Title: *Evaluation of the RIO-IFDM-street canyon model chain* Volume, pages: *77, 325-337*
2013	Environmental Science & Technology (IF 5.257)	Authors: *Kees de Hoogh, Meng Wang, Martin Adam, Chiara Badaloni, Rob Beelen, Matthias Birk, Giulia Cesaroni, Marta Cirach, Christophe Declercq, Audrius Dėdelė, Evi Dons, Audrey de Nazelle, Marloes Eeftens, Kirsten Eriksen, Charlotte Eriksson, Paul Fischer, Regina Gražulevičienė, Alexandros Gryparis, Barbara Hoffmann, Michael Jerrett, Klea Katsouyanni, Minas Iakovides, Timo Lanki, Sarah Lindley, Christian Madsen, Anna Molter, Gioia Mosler, Gizella Nador, Mark Nieuwenhuijsen, Goran Pershagen, Annette Peters, Harisch Phuleria, Nicole Probst-Hensch, Ole Raaschou-Nielsen, Ulrich Quass, Andrea Ranzi, Euripides Stephanou, Dorothea Sugiri, Per Schwarze, Ming-Yi Tsai, Tarja Yli-Tuomi, Mihaly J Varro, Danielle Vienneau, Gudrun Weinmayr, Bert Brunekreef, Gerard Hoek* Title: *Development of land use regression models for particle composition in 20 study areas in Europe* Volume, pages: *47 (11), 5778-5786*
2013	Atmospheric Environment (IF 3.110)	Authors: *Evi Dons, Martine Van Poppel, Bruno Kochan, Geert Wets, Luc Int Panis* Title: *Modeling temporal and spatial variability of traffic-related air pollution: Hourly land use regression models for black carbon* Volume, pages: *74, 237-246*
2013	Atmospheric Environment (IF 3.110)	Authors: *Rob Beelen; Gerard Hoek; Danielle Vienneau; Marloes Eeftens; Konstantina Dimakopoulou; Xanthi Pedeli; Ming-Yi Tsai; Nino Künzli; Tamara Schikowski; Alessandro Marcon; Kirsten Eriksen; Ole Raaschou-Nielsen; Euripides Stephanou; Evridiki Patelarou; Timo Lanki; Tarja Yli-Tuomi; Christophe Declercq; Grégoire Falq; Morgane Stempfelet; Matthias Birk; Josef Cyrys; Stephanie von Klot; Gizella Nádor; Mihály J Varró; Audrius Dėdelė; Regina Gražulevičienė; Anna Mölter; Sarah Lindley; Christian Madsen; Giulia Cesaroni; Andrea Ranzi; Chiara Badaloni; Barbara Hoffmann; Michael Nonnemacher; Ursula Krämer; Thomas Kuhlbusch; Marta Cirach; Audrey de Nazelle; Mark Nieuwenhuijsen; Tom Bellander; Michal Korek; David Olsson; Magnus Strömgren; Evi Dons; Michael Jerrett; Paul Fischer; Meng Wang; Bert Brunekreef; Kees de Hoogh* Title: *Development of NO_2 and NO_x land use regression models for estimating air pollution exposure in 36 study areas in Europe - the ESCAPE project* Volume, pages: *72, 10-23* Times cited: *3*
2013	Science of the Total Environment (IF 3.258)	Authors: *Evi Dons, Philip Temmerman, Martine Van Poppel, Tom Bellemans, Geert Wets, Luc Int Panis* Title: *Street characteristics and traffic factors determining road users' exposure to black carbon* Volume, pages: *447C, 72-79*

2012	Environmental Science & Technology (IF 5.257)	Authors: *Marloes Eeftens, Rob Beelen, Kees de Hoogh, Tom Bellander, Giulia Cesaroni, Marta Cirach, Christophe Declercq, Audrius Dėdelė, Evi Dons, Audrey de Nazelle, Konstantina Dimakopoulou, Kirsten Eriksen, Grégoire Falq, Paul Fischer, Claudia Galassi, Regina Gražulevičienė, Joachim Heinrich, Barbara Hoffmann, Michael Jerrett, Dirk Keidel, Michal Korek, Timo Lanki, Sarah Lindley, Christian Madsen, Anna Mölter, Gizella Nádor, Mark Nieuwenhuijsen, Michael Nonnemacher, Xanthi Pedeli, Ole Raaschou-Nielsen, Evridiki Patelarou, Ulrich Quass, Andrea Ranzi, Christian Schindler, Morgane Stempfelet, Euripides Stephanou, Dorothea Sugiri, Ming-Yi Tsai, Tarja Yli-Tuomi, Mihály J Varró, Danielle Vienneau, Stephanie von Klot, Kathrin Wolf, Bert Brunekreef, and Gerard Hoek* Title: *Development of land use regression models for $PM_{2.5}$, $PM_{2.5}$ absorbance, PM_{10} and PM_{coarse} in 20 European study areas: results of the ESCAPE project* Volume, pages: *46 (20), 11195-11205* Times cited: *5*
2012	Atmospheric Environment (IF 3.110)	Authors: *Evi Dons, Luc Int Panis, Martine Van Poppel, Jan Theunis, Geert Wets* Title: *Personal exposure to black carbon in transport microenvironments* Volume, pages: *55C, 392-298* Times cited: *8*
2011	Atmospheric Environment (IF 3.110)	Authors: *Evi Dons, Luc Int Panis, Martine Van Poppel, Jan Theunis, Hanny Willems, Rudi Torfs, Geert Wets* Title: *Impact of time-activity patterns on personal exposure to black carbon* Volume, pages: *45 (21), 3594-3602* Times cited: *23*
2011	Transportation Research Record: Journal of the Transportation Research Board (IF 0.442)	Authors: *Evi Dons, Carolien Beckx, Theo Arentze, Geert Wets, Luc Int Panis* Title: *Using an activity-based framework to determine effects of a policy measure on population exposure to Nitrogen Dioxide* Volume, pages: *2233, 72-79* Times cited: *3*

BOOK CHAPTERS

Year	Information
2012	Authors: *Evi Dons, Carolien Beckx, Theo Arentze, Geert Wets, Luc Int Panis* Chapter title: *Shop opening hours and population exposure to NO_2 assessed with an activity-based transportation model* Book title: Urban Environment. Proceedings of the 10th Urban Environment Symposium. Editors: *Sébastien Rauch; Gregory M Morrison* Pages: *161-170* ISBN: *978-94-007-2539-3*

LECTURES/PRESENTATIONS

Year	Information
2013	Title posters: - *Implementation and validation of a modeling framework to assess personal exposure to BC* - *HEAPS study design: Health Effects of Air Pollution in Antwerp Schools* Place: *Basel, Switzerland* Event: *Environment and Health. Conference of ISEE, ISES and ISIAQ*
2012	Title presentation: *Modeling exposure of schoolchildren to traffic-related air pollution: short term, medium term and long term exposure* (Presented by Luc Int Panis) Title poster: *New tools for integrated, personalized and dynamic exposure assessment* Place: *Antwerp, Belgium* Event: *BIOMAQ 2012: Biomonitoring of Air Quality*
2012	Title posters: - *Analysis of in-vehicle black carbon exposure and trip characteristics using GPS logs and diaries;* - *Trip motive in time and space: the impact on black carbon exposure* Place: *Seattle, USA* Event: *22nd Annual Meeting of the International Society of Exposure Science (ISES)*
2011	Title presentation: *Measuring personal exposure to black carbon: The importance of being in or near traffic* Place: *Barcelona, Spain* Event: *23rd Annual Conference of the International Society of Environmental Epidemiology (ISEE)*
2011	Title poster: *Impact of time activity patterns on personal exposure to black carbon* (Presented by Luc Int Panis) Place: *Boston, USA* Event: *Health Effects Institute, 2011 Annual Conference*
2011	Title presentation: *Personal monitoring of exposure to black carbon* Place: *Paris, France* Event: *Presentation due to an invitation of AirParif (Air quality monitoring in Paris)*
2011	Title poster: *Using an activity-based framework to determine the effects of a policy measure on population exposure to NO_2* (Presented by Carolien Beckx) Place: *Washington, D.C., USA* Event: *Transportation Research Board (TRB) 90th Annual Meeting*
2010	Title presentation: *Personal monitoring of exposure to black carbon* Place: *London, UK* Event: *Current & Future Air Quality Monitoring - Royal Society of Chemistry. Workshop and Conference with Posters and Exhibition. Jointly organized by the Automation and Analytical Management Group - Royal Society of Chemistry and AirMonTech*
2010	Title presentation: *Winkelopeningsuren en hun effect op de blootstelling van de bevolking aan NO_2* Place: *Roermond, The Netherlands* Event: *Colloquium Vervoersplanologisch Speurwerk 2010*
2010	Title presentation: *Shop opening hours and population exposure to NO_2 assessed with an activity-based transportation model* Place: *Ghent, Belgium* Event: *8th International Conference on Geostatistics for Environmental Applications*
2010	Title presentation: *Shop opening hours and population exposure to NO_2 assessed with an activity-based traffic model* Place: *Gothenburg, Sweden* Event: *10th Urban Environment Symposium*
2010	Title poster: *Widening shop opening hours and the effects on population exposure to NO_2* Place: *Antwerp, Belgium* Event: *2de Vlaams-Nederlandse Dag 'Milieu en Gezondheid'*

CONFERENCE ABSTRACTS

Evi Dons, Martine Van Poppel, Bruno Kochan, Geert Wets, Luc Int Panis (2013), Implementation and validation of a modeling framework to assess personal exposure to black carbon, Environment and Health - Conference of ISEE, ISES and ISIAQ.

Evi Dons, Martine Van Poppel, Sofie De Prins, Tim Nawrot, Luc Int Panis, Gudrun Koppen (2013), HEAPS study design: Health Effects of Air Pollution in Antwerp Schools, Environment and Health - Conference of ISEE, ISES and ISIAQ.

Evi Dons, Martine Van Poppel, Sofie De Prins, Gudrun Koppen, Christine Matheeussen, Tim Nawrot, Luc Int Panis (2012), Modeling exposure of schoolchildren to traffic-related air pollution: short term, medium term and long term exposure, Published in: Book of abstracts: BIOMAQ 2012 – Biomonitoring of Air Quality. p. 55-57

Evi Dons, Jan Theunis, Bart Elen, Martine Van Poppel, Patrick De Boever, Roel Smolders, Luc Int Panis (2012), New tools for integrated, personalized and dynamic exposure assessment, Published in: Book of abstracts: BIOMAQ 2012 – Biomonitoring of Air Quality. p. 83

Evi Dons, Philip Temmerman, Martine Van Poppel, Tom Bellemans, Geert Wets, Luc Int Panis (2012), Analysis of in-vehicle black carbon exposure and trip characteristics using GPS logs and diaries, Published in: The International Society of Exposure Science - Abstract Book. p. 19 + 161

Philip Temmerman, Evi Dons, Martine Van Poppel, Tom Bellemans, Geert Wets, Luc Int Panis (2012), Trip motive in time and space: the impact on black carbon exposure, Published in: The International Society of Exposure Science - Abstract Book. p. 107-108

Evi Dons, Martine Van Poppel, Jan Theunis, Geert Wets, Luc Int Panis (2011), Measuring personal exposure to black carbon: the importance of being in or near traffic, Published in: 23rd Annual Conference of the International Society for Environmental Epidemiology – Conference Abstracts.

Evi Dons, Luc Int Panis, Martine Van Poppel, Hanny Willems, Rudi Torfs, Geert Wets (2011), Impact of time-activity patterns on personal exposure to black carbon. Published in: Health Effects Institute – Annual Conference 2011. Program and Abstracts. p. 34.

Evi Dons, Carolien Beckx, Theo Arentze, Geert Wets, Luc Int Panis (2011), Using an activity-based framework to determine the effects of a policy measure on population exposure to NO_2, Published in: Proceedings DVD of the 90th Annual Meeting of the Transportation Research Board.

Evi Dons, Jan Theunis, Martine Van Poppel, Rudi Torfs, Luc Int Panis (2010), Personal monitoring of exposure to black carbon, Published in: Proceedings: Monitoring Ambient Air 2010 - New Air Quality Measurement Technologies. Current & Future Air Quality Monitoring

Evi Dons, Carolien Beckx, Luc Int Panis (2010), Winkelopeningsuren en hun effect op de blootstelling van de bevolking aan NO_2, Published in: Colloquium Vervoersplanologisch Speurwerk 2010. De stad van straks: Decor voor beweging. Programma en samenvattingen. p. 48

Evi Dons, Carolien Beckx, Theo Arentze, Geert Wets, Luc Int Panis (2010), Shop opening hours and population exposure to NO_2 assessed with an activity-based transportation model, Published in: 8th International Conference on Geostatistics for Environmental Applications. Book of Abstracts of geoENV 2010. p. 134-136. ISBN 978-94-9069-539-2

Evi Dons, Stijn Dhondt, Theo Arentze, Jan Theunis, Geert Wets, Carolien Beckx, Luc Int Panis (2010), Shop opening hours and population exposure to NO_2 assessed with an activity-based traffic model, Published in: 10th Urban Environment Symposium. Book of Abstracts. p. 31.

Evi Dons, Carolien Beckx, Luc Int Panis, Theo Arentze, Geert Wets (2010), Widening shop opening hours and the effects on population exposure to NO_2, Published in: Vlaams-Nederlandse CongresDag Milieu & Gezondheid. Visie, Gezondheidswinst en Toekomst. p. 56

AWARDS

Year	Country	Information
2012	USA	Title poster: *Analysis of in-vehicle black carbon exposure and trip characteristics using GPS logs and diaries* Description: *Student Poster Award 2012 – First place* Place: *Seattle* Institution: *22nd Annual Meeting of the International Society of Exposure Science (ISES)*
2010	Belgium	Title poster: *Widening shop opening hours and the effects on population exposure to* NO_2 Description: *Best Poster Award* Place: *Antwerp* Institution: *2de Vlaams-Nederlandse Dag 'Milieu en Gezondheid'*

TEACHING

- Hasselt University – Master Business Engineering: Guest lecture "Transport geography" (Academic year 2010-2011 and 2011-2012)
- Hasselt University – Supervising M.S. Candidate, Transportation Sciences (Philip Temmerman): "Analysis of the link between black carbon exposure and trip characteristics from GPS and time-activity diaries" (Academic year 2011-2012)

WORKSHOP PARTICIPATION

Year	Date	Country	Information
2012	Sept 21, 2012	Belgium	Title: *From PhD to Job Market / Workshop: Networking* Place: *Ghent* Description: *organized by the doctoral schools of Ghent University, Free University Brussels and Antwerp University*
2011	Sept 29, 2011	Belgium	Title: *Seminar: Introduction into R data management and statistics* Place: *Mol* Description: *VITO internal course*
2011	June 29, 2011	Belgium	Title: *GeoDa workshop: Spatial data analysis* Place: *Hasselt* Description: *Intensive (5-hours) GeoDa workshop*
2011	May 5 + May 12, 2011	Belgium	Title: *Seminar: Multi-level analysis* Place: *Hasselt* Description: *Hasselt University lunchseminar*
2010	Sept 11 – Sept 12, 2010	Belgium	Title: *Workshop: Introduction to geostatistical estimation and simulation* Place: *Ghent* Description: *pre-conference workshop "8th International Conference on Geostatistics for Environmental Applications"*
2010	April 26 – April 29, 2010	The Netherlands	Title: *Workshop Land Use Regression (LUR)* Place: *Utrecht* Description: *organized by IRAS (Institute for Risk Assessment Sciences – Utrecht University) in the framework of the FP7 project ESCAPE*

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