Measurement and modeling of short- and long-term commuter exposure to traffic-related air pollution

INAUGURALDISSERTATION

zur
Erlangung der Würde einer Doktorin der Philosophie
vorgelegt der
Philosophisch-Naturwissenschaftlichen Fakultät
der Universität Basel

von
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aus Flims (GR)

Basel, 2015

Genehmigt von der Philosophisch-Naturwissenschaftlichen Fakultät auf Antrag von Prof. Dr. Jürg Utzinger, Prof. Dr. Nino Künzli und Prof. Dr. Eberhard Parlow

Basel, den 15. Oktober 2013

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ACKNOWLEDGMENTS

This PhD work was part of the European wide project Transportation Air pollution and Physical ActivitieS: an integrated health risk assessment program of climate change and urban policies (TAPAS) and was financed by the Coca-Cola Foundation and the Swiss Tropical and Public Health Institute (Swiss TPH).

I dedicate this thesis to my first supervisor Lee-Jane Sally Liu who sadly passed away in June 2011. She strongly encouraged me, after doing my master's thesis with her, to become a researcher and do a PhD. I remember when I accepted her offer to do a PhD with her team, she was very happy, giving me a smile and a hug. This was a wonderful start, and I am deeply grateful to her for giving me the opportunity to go this way. I have truly learned from her never-failing motivation and her endless ideas and visions for research.

I like to thank the many people who have contributed to this PhD thesis in different ways. Above all, very special thanks to Harish Phuleria and Nino Künzli who took over the supervision of my PhD thesis. I am deeply thankful for their excellent advice, kind encouragement and on-going support.

I would like to thank the TAPAS team and the TAPAS partners from the six cities for their support during my PhD studies. Many thanks go to Charlotte Braun-Fahrländer. She always found time for me to discuss scientific and personal issues. I am also grateful to Audrey de Nazelle, the coordinator of the project, who was always ready to provide valuable input and connect me with needed experts across Europe and America. Thanks also go to Michael Jerrett from University of Berkley for his feedback and ideas.

Many thanks are addressed to Christian Schindler for statistical support. He was always available for help and ideas during the analysis and paper preparation.

I would like to thank the Environment Exposure and Health unit and former Environmental Exposure Science unit, as well as other colleagues at the Swiss TPH for supporting my work. Special thanks go to Ming-Yi Tsai, Alex Ineichen, Regina Ducret, Katrin Uehli, Elisabetta Corradi, Laura Perez and Martin Röösli. I want to further thank the PhD community at the Swiss TPH for scientific and motivational support.

I also would like to thank Mark Davy from the University of Washington in Seattle for teaching me organizational and technical issues of personal air pollution measurements and for his help in handling technical problems with the measurement devices. Thanks go also to all people helping me during the field work, data collection and data processing, namely

Alex Ineichen, Chantal Wullimann, Elisabetta Corradi, Damiano Urbinello and Siventher Jeyachchandren.

I am thankful to Jürg Jermann from the Civil Engineering Department Canton Basel-Land, Switzerland for his support related to Geographic Information Systems (GIS).

I wish to thank Harish Phuleria, Regina Ducret and Lisa Crump for proofreading my thesis and giving me helpful feedback. I also thank Noëmi Bachmann for the help in formatting my thesis and for designing the cover.

I am grateful to Prof. Dr. Eberhard Parlow for his support as co-referent and Prof. Dr. Jürg Utzinger from the Swiss TPH for attending this thesis as representative of the faculty.

I would like to express my deep gratitude to my family, especially to my parents for always believing in me and for supporting me in everything that I do. Last but not least, many thanks and all my love to Thomas. Thank you for the endless emotional support, encouragement and for being there for me.

Martina Ragettli

SUMMARY

Background

Many epidemiological studies have reported associations between traffic-related air pollution exposure and acute and chronic health problems. Exposure assignment in those studies has typically relied on home outdoor locations and ignored exposure during commuting and at non-residential locations. However, because of high concentrations of harmful air pollutants in proximity to traffic, time spent in transport may contribute considerably to a person's total daily exposure to traffic-related air pollution. An understanding of how activity patterns affect exposure to traffic-related air pollution in space and time is important for improved exposure assessments.

Concentration levels and individuals' exposures to harmful traffic-related air pollutants in the various transport microenvironments are not well understood. Recently, exposure to ultrafine particles (UFP, particles smaller than 100 nm) has attracted particular interest. UFP are considered harmful to human health in view of their small size and the probability to penetrate deeply into the respiratory tract. Little is known about the variability in UFP concentrations and most notably the average particle size in various transport environments. This is largely due to the lack of a robust portable device to measure UFP characteristics.

Objectives

The aim of this thesis was to characterize exposure to both UFP concentration and average particle size distribution diameters in commonly used transport environments in Basel. In addition, a simulation of commuter exposure to traffic-related air pollution of a general population was carried out to estimate the contribution of commute (i.e., the time spent in traffic traveling between home and work or school) to total exposure and inhalation dose as well as its relevance in epidemiological studies on long-term health effects of traffic-related air pollution.

Methods

Three sub-studies were performed to characterize personal exposure to UFP concentration and average particle size distribution diameters in frequently traveled commuter microenvironments. The personal monitoring campaign was carried out in the city of Basel and surrounding area between December 2010 and September 2011 using a newly developed portable device, the miniature Diffusion Size Classifier (miniDiSC), which measures particles

in the size range of 10 to 300 nm. First, the spatial variation of sidewalk UFP exposures within urban areas and transport-specific microenvironments was explored. Measurements were conducted along four predefined walks once per month. Second, exposure to UFP concentration and average particle size were quantified for five modes of transportation (walking, bicycle, bus, tram, car) during different times of the day and week, along the same route. Finally, the contribution of bicycle commuting along two different routes (along main roads, away from main roads) to total daily exposures was assessed by 24-hour personal measurements. Measurements were equally distributed over weekdays (Monday to Friday) across three seasons – winter, spring and summer.

The simulation of commuter exposure to traffic-related air pollution was conducted based on spatially and temporally resolved data on commuter trips of residents working (or attending a school) within the Basel area (Cantons Basel-City and Basel-Country). The information on commuter routes, transportation modes and home, work and school locations were extracted from the year 2010 Swiss Mobility and Transport Microcensus survey. An approach to simulate travel routes based on the transportation mode and origin/destination location of the legs (pieces of the trips with the same transportation mode) was developed and validated. Individuals' exposures to NO_2 during commuting and at home, work and school locations were computed by overlapping the locations and travel routes with annual mean maps of NO_2 in a geographic information system (GIS). Three air pollution models (a land use regression model (LUR), a high and a low resolution dispersion model) were evaluated for estimating commuter exposures to NO_2 as a marker of long-term exposure to traffic-related air pollution. Finally, the bias in health effect estimates resulting from using home outdoor exposures only and ignoring other non-residential exposures including commuter exposure was quantified.

This thesis is part of the Europe-wide project, Transportation Air Pollution and Physical ActivitieS (TAPAS), which is an integrated health risk assessment program on climate change and urban policies.

Results

In general, smaller average particle sizes and higher UFP concentration levels were measured at places and for transportation modes in close proximity to traffic. Average trip UFP concentrations were highest in car (31,800 particles cm^{-3}) followed by bicycle (22,700 particles cm^{-3}), walking (19,500 particles cm^{-3}) and public transportation (14,100-18,800 particles cm^{-3}). Concentrations were highest for all transportation modes during weekday morning rush hours, compared to other time periods. UFP concentration was lowest in bus, regardless of time period. Average particle diameters followed an opposite trend than UFP concentration, showing larger average particle sizes for transportation modes and sampling times with lower UFP number concentrations and vice versa. Bicycle travel along main streets between home and work place (24 min on average) contributed 21% and 5% to total

daily UFP exposure in winter and summer, respectively. Contribution of bicycle commutes to total daily UFP exposure could be reduced by half if main roads were avoided.

Within Basel-City, estimated average time-weighted NO_2 population exposure during commuting was similar among all air pollution models (around 39-41 µg m^{-3}). The spatial variability in NO_2 concentrations, as typically encountered in urban street environments, was best reflected by the dispersion model with the highest resolution (grid size of 25 m). By comparison, both the LUR model (applied to a 50x50 m grid) and the dispersion model with a lower resolution (100x100 m) underestimated the NO_2 concentrations on the higher end, and overestimated the values on the lower end.

The population working (>= 50% work load) or attending a school within the region of Basel spent on average 49 minutes for daily commutes. Work or school occupied 22% of the subjects' time on average. Median contribution of commuting to total weekly NO_2 exposure was 2.7% (range 0.1-13.5%). With regard to inhalation dose, the commute contributed slightly more when assuming moderate (3.5%, range: 0.2-16.8%) or high (4.2%, range: 0.2-33.0%) breathing rates during active transportation. The median contribution of commute to the total NO_2 exposure was highest for subjects using mainly public transportation (4.7%, range: 1.3-13.5%) who also spent the longest time in traffic (more than an hour). The comparison between the transportation modes based on the legs of the trips, however, revealed the highest NO_2 exposures for motorized transportation.

The failure to differentiate between outdoor NO_2 exposure at work/school and at home could result in a 12% (95%-CI: 11-14%) underestimation of related health effects. This bias was stronger for the subjects commuting between Basel-City and the rural to suburban surrounding areas of Basel-Country (33% underestimation) than for the subjects commuting within those areas. For the same population sub-group, potentially significant underestimation of health effects (5%, 95%-CI: 4-5%) attributable to including outdoor exposures at home and at work/school but omitting exposure during the commute was found.

Conclusions and outlook

This thesis provides important insights in the spatial and temporal variability of UFP within an urban area and provides an approach for modeling commuter exposures to traffic-related air pollution in epidemiological studies. Results confirmed the expectation that people are exposed to potentially high exposures during their daily travels and that ignoring time-activity patterns in epidemiological studies results in exposure misclassification and bias associated health effects.

The benefit of incorporating non-residential locations and daily commute patterns in exposure assignments of future epidemiological studies should carefully be evaluated based on (1) spatial and temporal variability of the pollutants of interest, and (2) the spatial spread of

home and work/school locations and subjects' level of mobility. Improved exposure estimation thus requires information on subjects' travel duration, distance, transportation modes, trip timings, route choices and work load.

Future exposure assessments of large cohorts will need to more frequently combine modeling approaches with actual personal exposure measurements of pollutants of interest to refine and validate exposure estimates spatially as well as temporally.

LIST OF ABBREVIATIONS AND DEFINITIONS

BC	Black carbon
CI	Confidence interval
CNG	Compressed natural gas
CPC	Condensation particle counter
CO_2	Carbon dioxide
CO	Carbon monoxide
GIS	Geographic information system
GPS	Global positioning system
I/O ratio	Indoor/outdoor ratio, or in-cabin/on-road ratio
LUR	Land use regression model
miniDiSC	Miniature Diffusion Size Classifier
NO	Nitric oxide
NO_2	Nitrogen dioxide
NO_x	Nitrogen oxides (includes NO and NO_2)
O_3	Ozone
PM	Particulate matter
$PM_{2.5}$	Particulate matter, particles with an aerodynamic diameter ≤2.5 μm
PM_{10}	Particulate matter, particles with an aerodynamic diameter ≤10 μm
SD	Standard deviation
TAPAS	Name of the research project; Transportation, Air pollution and Physical ActiviteS
UFP	Ultrafine particles, particles with an aerodynamic diameter <100 nm
VOC	Volatile organic compounds

1 INTRODUCTION AND BACKGROUND

1.1 Preface

Air pollution is a major environmental and public health problem. Research in recent decades has consistently shown adverse effects of outdoor air pollution on human health. In 2010, outdoor air pollution was ranked among the ten most important risk factors attributed to the global health burden, being responsible for approximately 3.2 million deaths worldwide per year (Lim et al., 2012). In urban areas, road traffic is one of the most important sources of ambient air pollution, and therefore an important contributor to the many well established health effects associated with general urban air pollution. There is sufficient scientific evidence that near-road traffic-related air pollution causes specific health effects that may partly occur independent of the background air pollution mixtures, such as exacerbation of asthma, various respiratory symptoms, impaired lung function and cardiovascular mortality and morbidity (HEI, 2010).

Research efforts on near-road traffic-related air pollution exposure levels and the relationships to human health have mainly focused on home environment settings. Traditional exposure assessment approaches do not take into account commuter behavior but rely mostly on home addresses with the assumption that people spend most of their time at home. This thesis focuses on in-traffic air pollution exposures of individuals and the general population. Because of high concentrations of harmful air pollutants, proximity to traffic, and because many journeys are made during rush hours, time spent in transport may contribute considerably to a person's total daily exposure to air pollution (WHO, 2006). To study people's exposure to near-road traffic-related air pollution, one must be aware of the specific pollutants and their spatial variability.

1.2 In-transit air pollution

Traffic-related air pollution is a complex mixture of various gaseous compounds and particulates. Vehicle exhaust produced by fuel combustion contains a range of potentially harmful pollutants, including carbon monoxide (CO), nitrogen oxides (NO_x), including nitric oxide (NO) and nitrogen dioxide (NO_2), volatile organic compounds (VOC) and particulate matter (PM). Additional so-called non-exhaust pollutants arise from the abrasion of tires and brake linings and resuspended dust from road surfaces. Air pollution due to vehicle traffic is also secondarily formed through physical and chemical processes (e.g. NO_2, secondary gases and aerosols such as ozone, nitrates, sulfates).

Traffic-related PM exists as liquids, solid or semivolatile components covering a wide range of sizes. Generally, PM is divided in *coarse* (particles with an aerodynamic diameter of 2.5-10 µm), *fine* (particles less than 2.5 µm or $PM_{2.5}$) and *ultrafine* (particles smaller 0.1 µm or smaller 100 nm) size fractions. Traffic-related coarse particles originate mainly from non-exhaust emissions, while fine and ultrafine particles are formed by vehicular exhaust emissions. In urban environments, ultrafine particles (UFP) constitute up to 95% of the total number concentration but contribute little to particle mass (Morawska et al., 2008). Therefore, UFP are usually measured as number counts per unit volume of air, whereas PM_{10} and $PM_{2.5}$ is reported in terms of mass concentration.

The size of PM also indicate how deeply inhaled particles penetrate into the human respiratory tract. Moreover, particles can carry other substances on their surfaces, such as oxidant gases, organic compounds and transition metals, of which some are toxic or carcinogenic. Smaller particles provide a larger surface area to carry such chemicals (Valavanidis et al., 2008). PM_{10} can penetrate into the finest branches of the bronchial system. Particles less than 2.5 µm are sufficiently small to enter the lungs and may even reach the alveoli. UFP are considered most harmful in view of the probability to penetrate deeply into the alveolar region, to bridge tissue barriers and to adsorb and retain toxic substances (Peters et al., 2011; Rueckerl et al., 2011; WHO, 2006).

The pollutant mixture originating from vehicles can vary and decrease rapidly in concentration within short distances away from traffic. In particular, UFP have a relatively short lifetime in the urban atmosphere as they tend to aggregate to form larger particles. UFP have been shown to decrease by 30% at 40 meter distance from the highway and by 70% at 100 meter distance (Zhu et al., 2002). Coagulation processes reduce the number concentrations and shift the size distribution to larger sizes, thus, number concentrations of fine and coarse particles show less spatial variability in the vicinity of the road than ultrafine particles. Other gaseous co-pollutants such as NO_2 and CO also show rapid decrease with distance away from the road, although with smaller decay gradients, reaching background levels at around 200 to 500 m (Beckerman et al., 2008; Zhou and Levy, 2007). Pollutants that show a sharp decrease with short distance from the road serve as indicator pollutants for near-road traffic-related (primary) air pollutants. However, their high variability within a few meters away from the roadway, especially for UFP, poses challenges to characterize both the spatial and temporal concentration gradients within an urban area.

1.3 Methods of exposure assessment to traffic-related air pollution

1.3.1 The concept of exposure assessment

Monitoring human exposure to traffic-related air pollution is complex not only due to the spatial and temporal variability of traffic pollutants, but also due to the fact that traffic-related pollution consists of a variety of pollutants which all originate also from other sources such as for example from domestic heating or industry (HEI, 2010).

The basic concept of exposure assessment, i.e. the pathway of how a pollutant of a given source may lead to a human health response, is illustrated in Figure 1-1. In exposure science, air pollution is often characterized by an indicator pollutant (also called a marker or tracer) of a *source* of interest. Commonly used tracer pollutants for fresh traffic exhaust are NO_2, CO, benzene (a carcinogenic VOC), PM mass – in particular the black carbon (BC) fraction (dark, light-absorbing component of PM mainly from diesel fuels) – and UFP. *Concentrations* of such pollutants depend on the emission strength of the source and the composition of the vehicle fleet (gasoline and diesel). The concentration levels are determined by factors affecting the dispersion, including meteorological conditions (e.g. wind speed, wind velocity, humidity), topography and characteristics of the built environment (e.g. building density, building height) (HEI, 2010; Knibbs et al., 2011; Vardoulakis et al., 2003). "The event when a person comes into contact with a pollutant of a certain concentration during a certain period of time" (Ott, 1982, p. 186) is defined as the *personal exposure*. Therefore, high concentration levels do not necessarily mean high exposure if only a short time interval is spent at such areas. The *inhaled dose* refers to the amount of pollutants absorbed or deposited in the human body during a period of time which may lead to a *health effect*. The biologically relevant dose depends on physical and chemical properties of the pollutants (e.g. efficiency of deposition in the respiratory tract) as well as on physiological characteristics such as breathing frequency and tidal volume, thus, on the physical effort (Hofmann, 2011).

Figure 1-1. The concept of air pollution exposure (adapted from WHO 2006, p. 62)

1.3.2 Assessment of exposure to traffic-related air pollution

In epidemiological studies, exposure assessment of a specific indicator pollutant typically relies on fixed-site measurements within a city, as often provided from routine air quality monitoring. Those measurements serve primarily to comply with air pollution standards and regulations and to obtain information on the temporal trends on the urban scale. For pollutants with a high spatial and temporal variability, these local estimates of exposure are, however, representative neither for the whole population nor for all traffic micro-environments (HEI, 2010). Consequently, exposure assignment has focused more on the local differences, thus on within-area and between-subject variability. As it is not feasible in studies with large sample sizes to measure individual traffic exposure for all participants, some studies used measures of traffic itself such as distance between home locations and major roads (Bayer-Oglesby et al., 2006; Hazenkamp-von Arx et al., 2011; McConnell et al., 2006; Tonne et al., 2007; Venn et al., 2001), traffic intensity or traffic density around locations of interest (Brunekreef et al., 1997; Nicolai et al., 2003; Venn et al., 2000). However, such parameters may misclassify exposures as they are not based on actual air pollution monitoring data (Jerrett et al., 2005).

Recently, various model approaches have been applied in epidemiological studies to better capture the spatial variability of air pollutants. Such model techniques include geo-statistical interpolation techniques of fixed-site data, dispersion models that use emission data from several sources and meteorological parameters and land use regression (LUR) models. The latter incorporate land use information and traffic data in addition to fixed site measurements to predict air pollution concentration for an area and locations of interest. With such techniques, spatial maps for a given air pollutant and a certain area, typically for annual average concentration, are computed and allow for spatially assigning the concentration to a specific population of interest.

These methods generally assume exposure to equal the outdoor air pollution concentration of a person’s residence, census tract or postal code, while ignoring individual mobility patterns. To date, only a few attempts (e.g. Beckx et al., 2009; Marshall et al., 2006; Setton et al., 2008) have been made to model exposure to air pollution in traffic for a large population and its sub-groups. This can be explained by the fact that modeling journey-time exposure is very difficult due to the spatial and temporal dynamics of both the population and air pollution concentrations (Gulliver and Briggs, 2005; Jerrett et al., 2005).

1.3.3 Personal in-traffic exposure assessment

Exposure can also be directly measured by means of personal monitoring which utilizes a portable device, ideally in the breathing zone, for a certain time to assess an individual’s exposure to a pollutant. Personal monitoring studies provide important insights of exposure characteristics and determinants of a given transport environment. This direct approach is considered most accurate when investigating the actual exposures of people during their daily activities (Steinle et al., 2013). It has been used to compare people’s actual exposures with static concentration data from fixed-site network stations and to validate air pollution models.

Personal monitoring studies, however, normally provide only limited information on the commuter exposure levels of an entire urban population. Exposure estimates are related to a small number of individuals over short time intervals. Usually, in-transit measurements are carried out along predefined routes rather than routes representing real population-based activity patterns. In addition, personal monitoring has generally been not feasible for large cohort studies due to the high costs and the commitments requested from study participants (Steinle et al., 2013).

The recognition that commuters' exposures are both highly elevated when compared to elsewhere and potentially harmful emerged in the 1960s, when the first CO measurements in cars on heavily trafficked Los Angeles roads were carried out by Haagen-Smit (Haagen-Smit, 1966). In recent years, a growing number of studies exploring the levels and determinants of air pollution exposure in traffic have been published. In-vehicle exposure levels have been studied most extensively, while walking and cycling have been less frequently included (Kaur et al., 2007; Knibbs et al., 2011). Few multi-modal studies (e.g. Kaur et al., 2005; McNabola et al., 2008) including four or more transportation modes have been conducted. The attention shifted from gaseous compounds such as CO and VOC to PM, and most recently – with the notion of being a public health concern and the development of appropriate monitoring devices – to UFP and black carbon. Air pollution exposure levels of cyclists and pedestrians have generally been reported to be lower than for occupants of cars and buses (Kaur et al., 2007; Knibbs et al., 2011), except for one study where higher exposure levels were found for walking than in car (Briggs et al., 2008). In addition, the majority of existing studies addressing UFP has focused on particle number concentration and not on particle size distribution in different transport environments, which is largely due to the lack of portable devices to measure particle size.

1.4 Policies to reduce traffic-related air pollution

With the recognition of traffic-related air pollution being a public health risk, various actions on national and international levels have been taken to improve air quality and ensure human well-being. Legislation to reduce tailpipe emissions were released which was/is achieved by development of cleaner fuels, after-treatment technology and newer engines. Many countries have defined air quality guidelines and standards for traffic-related air pollutants that are continuously evaluated at city-centre stations or national monitoring networks. For example, authorities in Europe and in the United States of America have set limiting values of various pollutants including PM_{10}, $PM_{2.5}$, ozone, CO, and NO_2. However, there is currently no ambient air quality standard for UFP because there is no standardized sampling procedure (Morawska et al., 2008) and no established exposure-response relationship. It is still an open question which metric is best for characterizing the toxicity of UFP. Particles may pose different health risks depending on their properties, namely, number concentration, particle size, shape, surface area and chemical composition (Heal et al., 2012; Rueckerl et al., 2011). Current legislation in Europe focuses on limiting the emission of UFP by vehicle emission standards. International and national groups such as the World Health Organization regularly evaluate current knowledge from epidemiological studies and present conclusions for further actions. Additional measures to improve air quality include the promotion of transportation modes that are safer for health and the environment. This encompasses transportation and spatial planning policies promoting walking, cycling and public transportation as alternatives to using private cars (WHO, 2005).

1.5 Rationale

Exposure assignment in epidemiological studies on the long-term health effects of traffic-related air pollution has mostly relied on home outdoor locations and ignored the potential impact of individual mobility patterns such as time spent in transport and at work. Concentration levels and individuals' exposures to harmful traffic-related air pollutants within the city and in the various transport microenvironments are not well understood. Especially in regard to UFP, this gap is mainly due to limitations of fixed-site air quality monitoring networks to provide information on individual exposure and unavailability of accurate portable measurement devices. Improved approaches for modeling in-transport exposure to traffic-related air pollution for a large number of individuals are needed. An understanding of how activity patterns affect exposure to traffic-related air pollution in space and time is important for improved exposure assessments. It will further help in the evaluation and elaboration of urban policies addressing public health and transport management.

2 FRAMEWORK AND OBJECTIVES

2.1 The TAPAS project

This thesis work is part of the European wide project **T**ransportation **A**ir pollution and **P**hysical **A**ctivitie**S**: an integrated health risk assessment program of climate change and urban policies (**TAPAS**). The purpose of the project was to help decision makers design urban policies that address climate change and promote good health. In particular, the work included assessments of conditions and policies that hinder or encourage active travel. The aim of the TAPAS research programme was to assess health impacts of active transport policies in an integrated framework in six case-study cities: Barcelona, Basel, Copenhagen, Paris, Prague, and Warsaw.

The underlying idea is that shifting the population towards active transportation (i.e. promoting walking and using the bicycle) may address some of the greatest public health challenges, such as urban air pollution, climate change mitigation through reduced carbon emissions (Woodcock et al., 2009) or physical inactivity (Frank et al., 2006). However, modal shifts from motorized to non-motorized transportation may also result in negative health effects among those physically active in urban streets, as for example through increased inhalation of air pollution and increased accident rates (de Nazelle et al., 2011).

As a first step to achieve the aims, a conceptual framework characterizing potential risks and benefits of interventions that promote active travel was developed. The framework was developed in workshops with experts from various related fields of research. Secondly, quantitative models of impacts of active travel policies were built. Input data as well as policy examples were provided from the TAPAS case cities. The aim was also to involve local stakeholder to identify local needs and produce local interest.

The TAPAS project was coordinated by the Centre for Research in Environmental Epidemiology (CREAL) in Barcelona. The development of the conceptual framework and the quantitative model development were led by CREAL. The TAPAS case-studies developed specific research projects to provide new knowledge filling research gaps in the framework. The focus of the TAPAS Basel project was to assess in-transit exposures, so-called commuter exposure, to traffic-related air pollution.

2.2 Aims of this thesis

The overarching aim of this thesis was to get a better understanding of commuter exposure to traffic-related air pollution. This is considered important for epidemiological health assessment studies as well as for the enactment and implementation of public health and transport policies. In particular, the aims were **first**, to assess personal commuter exposure to ultrafine particles – including both particle number and average particle size – in various traffic environments in Basel; **second**, to model population commuter exposure and to assess the applicability of different air pollution models in estimating commuter exposure to NO_2 in epidemiological studies; and **finally** to assess the contribution of commute to total NO_2 exposure of individuals in a representative population sample and to investigate its relevance in epidemiological studies on the long-term health effects of traffic-related air pollution.

The specific aims and various research questions addressed within are as follows:

Aim 1:	Characterize personal exposure to both ultrafine particle concentration and average particle size distribution diameters in commonly used transport microenvironments.

Specific aims:

I. Explore the spatial variation in UFP exposures within and between urban areas and transport-specific microenvironments.
II. Quantify UFP concentration and average particle size differences among five modes of transport (walking, bicycle, bus, tram, car) during different times of the day and week.
III. Study the contribution of bicycle commuting along potentially high and low exposure routes to total daily UFP exposure.

Aim 2:	Simulation of commute exposure to traffic-related air pollution of a general population.

Specific aims:

I. Estimate individual NO_2 exposures in a representative population sample during commute within the metropolitan area of Basel, Switzerland.
II. Evaluate the applicability of air pollution models with different spatial resolution and methodology to estimate commuter exposure and their applicability in long-term exposure assessment.

Aim 3:	Assessment of the contribution of commute to total exposure and inhalation dose and its relevance in epidemiological studies on long-term health effects of traffic-related air pollution.

Specific aims:

I. Investigate the contribution of the commute (i.e. the time spent in traffic travelling between home and work or school) to total NO_2 exposure and inhalation dose.

II. Quantify the potential bias expected in health effect estimates that can occur when outdoor pollution levels at home are used as estimates of total exposure and outdoor exposures at work or school and during commuting are ignored.

2.3 Outline of the thesis

Following the introduction and background (*chapter 1*) and aims (*chapter 2*), in *chapter 3*, the results from an extensive personal measurement campaign assessing ultrafine particles are presented (article 1 *"Commuter exposure to ultrafine particles in different urban locations, transport modes and routes"*). Personal monitoring was carried out in Basel, Switzerland, from December 2010 to September 2011.

In *chapter 4*, the results of modeling the long-term commuter exposure to traffic-related air pollution of the population are presented. First, the methods and results of the *simulation of population-based commuter exposure to* NO_2 *using different air pollution models* (article 2) are described and discussed. Second, the simulated NO_2 estimates are used to assess how much commute contributes to the total exposure and inhalation dose (article 3 *"The relevance of commuter and work/school exposure in an epidemiological study on traffic-related air pollution"*).

Finally, the main findings presented in chapters 3 to 4 are summarized in *chapter 5* and discussed in *chapter 6*. Strengths and limitations of the methodology are discussed. Further, scientific and policy implications and recommendations for future studies are elaborated.

3 MEASUREMENT OF PERSONAL COMMUTER EXPOSURE TO TRAFFIC-RELATED AIR POLLUTION

Article 1: Commuter exposure to ultrafine particles in different urban locations, transport modes and routes

This paper has been published:

Ragettli M. S., Corradi E., Braun-Fahrländer C., Schindler C., de Nazelle A., Jerrett M., Ducret R. E., Künzli N., Phuleria H. C., 2013. Commute exposure to ultrafine particles in different urban locations, transportation modes and routes. Atmospheric Environment 77, 376-384.

Atmospheric Environment 77 (2013) 376–384

Contents lists available at SciVerse ScienceDirect

Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv

Commuter exposure to ultrafine particles in different urban locations, transportation modes and routes

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HIGHLIGHTS

- UFP concentration and mean particle size were measured in transport environments.
- Personal sampling included urban areas, five travel modes and two bicycle routes.
- Highest UFP concentrations were measured in car and on bicycle, lowest in bus.
- Bicycle commutes contributed notably (21% in winter) to 24-h UFP exposure.
- Avoiding main roads reduced the contribution of bicycle commutes to total daily UFP.

ARTICLE INFO

Article history:
Received 10 October 2012
Received in revised form
2 May 2013
Accepted 3 May 2013

Keywords:
Air pollution
Ultrafine particles
Commuter
Near-road exposure
Particle size
Switzerland

ABSTRACT

A better understanding of ultrafine particle (UFP) exposure in different urban transport microenvironments is important for epidemiological exposure assessments and for policy making.

Three sub-studies were performed to characterize personal exposure to UFP concentration and average particle size distribution diameters in frequently traveled commuter microenvironments in the city of Basel, Switzerland. First, the spatial variation of sidewalk UFP exposures within urban areas and transport-specific microenvironments was explored. Second, exposure to UFP concentration and average particle size were quantified for five modes of transportation (walking, bicycle, bus, tram, car) during different times of the day and week, along the same route. Finally, the contribution of bicycle commuting along two different routes (along main roads, away from main roads) to total daily exposures was assessed by 24-h personal measurements.

In general, smaller average particle sizes and higher UFP levels were measured at places and for travel modes in close proximity to traffic. Average trip UFP concentrations were higher in car (31,784 particles cm^{-3}) and on bicycle (22,660 particles cm^{-3}) compared to walking (19,481 particles cm^{-3}) and public transportation (14,055–18,818 particles cm^{-3}). Concentrations were highest for all travel modes during weekday morning rush hours, compared to other time periods. UFP concentration was lowest in bus, regardless of time period. Bicycle travel along main streets between home and work place (24 min on average) contributed 21% and 5% to total daily UFP exposure in winter and summer, respectively. Contribution of bicycle commutes to total daily UFP exposure could be reduced by half if main roads are avoided.

Our results show the importance of considering commuter behavior and route choice in exposure assessment studies.

1. Introduction

Ultrafine particle (UFP, <100 nm) concentrations are usually particularly high along busy roads, common in urban transport environments (Morawska et al., 2008; Zhu et al., 2002). UFPs are

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http://dx.doi.org/10.1016/j.atmosenv.2013.05.003

generated in large quantities by fuel combustion processes, with vehicular traffic exhaust being the predominant source in urban environments (Morawska et al., 2008). Near-road UFP investigations have documented direct dependence of UFP levels on various traffic parameters such as distance to road (Buonanno et al., 2009; Zhu et al., 2002), traffic volume (Boarnet et al., 2011; Briggs et al., 2008; Kaur and Nieuwenhuijsen, 2009), and composition of the fleet (gasoline, diesel) (Fruin et al., 2008).

UFP exposure in transport environments has attracted interest since there is increasing evidence that ambient UFP exposure is associated with adverse health effects (Hoek et al., 2010). Toxicological and laboratory studies have demonstrated cardiovascular and respiratory health effects of UFP, which likely have different and partly independent effects from larger particles, due to their small size, large surface area, different chemical composition and ability to penetrate deep into the alveolar region and tissue barriers (Valavanidis et al., 2008). On average, people in Europe spend just 8% of their time in transport environments (Hänninen et al., 2005), yet even these limited windows may contribute considerably to their total UFP exposure (de Nazelle et al., 2012; Fruin et al., 2008; Zhu et al., 2007).

Assessing individual and population exposure to UFP in urban transport environments is challenging, as UFP concentrations and particle sizes vary within short distances from the source. Coagulation and atmospheric dilution processes contribute to a rapid decline in UFP concentration and increase of mean particle size distribution diameter within the first few meters away from the roadside (Buonanno et al., 2009; Zhu et al., 2002). As a result, fixed site monitors generally underestimate commuter exposures, especially on or near heavily traveled roads (Knibbs et al., 2011).

Additional factors may affect in-transit UFP exposures, like the mode of transport (de Nazelle et al., 2012; Kaur and Nieuwenhuijsen, 2009; Knibbs and de Dear, 2010), route (Zuurbier et al., 2010), vehicle configuration (Zuurbier et al., 2010) and commute duration (Briggs et al., 2008). These factors may vary considerably across geographical areas due to different meteorology, traffic characteristics and travel behavior. While several studies reported higher UFP levels in cars compared to other transportation modes (de Nazelle et al., 2012; Knibbs et al., 2011), Briggs et al. (2008) reported higher UFP levels for walking than car. Only a few UFP commuter exposure assessments compared more than two travel modes (de Nazelle et al., 2012; Kaur et al., 2005; Knibbs and de Dear, 2010; Zuurbier et al., 2010). The majority of existing commuter exposure studies has focused on particle number concentration and not on particle size distribution in different urban transport environments; this is largely due to the lack of a portable robust device to measure particle size characteristics.

To estimate long-term UFP commuter exposure in large populations, it is necessary to understand in-transit exposures within a given urban area. Thus, the present study characterizes personal exposure to both UFP concentration and average particle size distribution diameters in commonly used commuter microenvironments in Basel, Switzerland. The specific aims of this work were (1) to explore the spatial variation in UFP exposures within and between urban areas and transport-specific microenvironments, (2) to assess UFP concentration and average particle size differences among five modes of transportation (walking, bicycle, bus, tram, car) during different times of the day and week, and (3) to study the contribution of bicycle commuting along two different routes to total daily exposure.

2. Methods

2.1. Study design and location

Personal UFP measurements were carried out in the city of Basel and surrounding area between December 2010 and September 2011. The city, located in the Rhine valley (260 m above sea level), has about 190,000 inhabitants and has average temperatures of 3 °C–6 °C in winter, and 21 °C–25 °C in summer. Residents primarily use public transport (52%), private car (18%), or bicycle (17%) for their daily commute to work. In Basel, the fleet of vehicles is composed of 1% heavy duty vehicles, and 18% of the passengers cars are diesel (Cantonal Office of Statistics Basel-City, 2010).

The study was divided into three separate sub-studies: the *first* was conducted in and around Basel to assess spatial variation of UFP concentrations near roads and in different commuter microenvironments; the *second* quantified UFP levels for five different transportation modes on a major street in central Basel; and the *third* measured 24-h personal UFP concentrations, which included a potentially high and a low exposed bicycle commuter route between home and work. Routes and sampling locations are shown in Fig. 1. The details of each sub-study are described below.

2.2. Sub-study 1: spatial variation of near-road UFP

A walking route was defined in each of four urban areas – Basel city center (*Basel city center, pedestrian*), a green residential area on a hill (*Bruderholz, residential green*, 340 m above sea level), a densely populated residential area (*Gundeldingen, residential urban*) and a town center in the Basel metropolitan area (*Liestal town center, traffic*, 13,900 inhabitants) 13 km from Basel. Average vehicles/day along the routes were 700, 1200, and 7000 in Basel city center, *Bruderholz* and *Gundeldingen*, respectively (Department of Public Works and Transport Canton Basel-City, 2008). The Basel city center is characterized by a mix of pedestrian zones and streets with limited traffic. Buildings typically have three to five stories. In addition, measurements were conducted at five non-sheltered bus and tram stops in *Gundeldingen*, Basel city center, and in front of the Basel main train station along or before/after the walk. Each walk was conducted one after another on the first Wednesday or Thursday morning (8am–12pm) of each month. Monitoring was similarly conducted inside the main train station (on platforms and in waiting areas), in an underground bicycle parking garage, inside the EuroAirport Basel and inside two main shopping centers in Basel city center on either or both of these two days.

2.3. Sub-study 2: UFP in different transportation modes

Repeated measurements were carried out for five transportation modes (walking, bicycle, bus, tram, car) on the same route along a main road in Basel (see Appendix A for more details on route characteristics). Samples were collected during 13 days in spring (March 22nd–May 18th) and five days in fall (September 21st–28th). Days with similar weather conditions (no rain, similar temperatures) were chosen for monitoring. Samples were collected at three time periods characterized by similar traffic conditions: weekday rush hour (7–9am, 4:30–6:30pm), weekday non-rush hour (10am–11:30am, 2–3:30pm) and on the weekend (10am–3pm). Weekdays were restricted to Tuesday through Thursday, while weekend included both Saturdays and Sundays. For weekday measurements, samples were collected consecutively at least four times (twice in each direction) for each mode of transport and each time slot. Weekday rush and non-rush hours were split into morning and afternoon. For practical reasons, weekday measurements for walking, cycling, bus and tram trips were made on the same day, while measurements for car trips were conducted on the day before or after. Weekend measurements covered all five modes on the same day.

Buses were powered by diesel (equipped with particulate filters) or compressed natural gas (CNG) and were mechanically ventilated with windows closed. The electrically powered high-floor trams (1990s model) were not equipped with mechanical

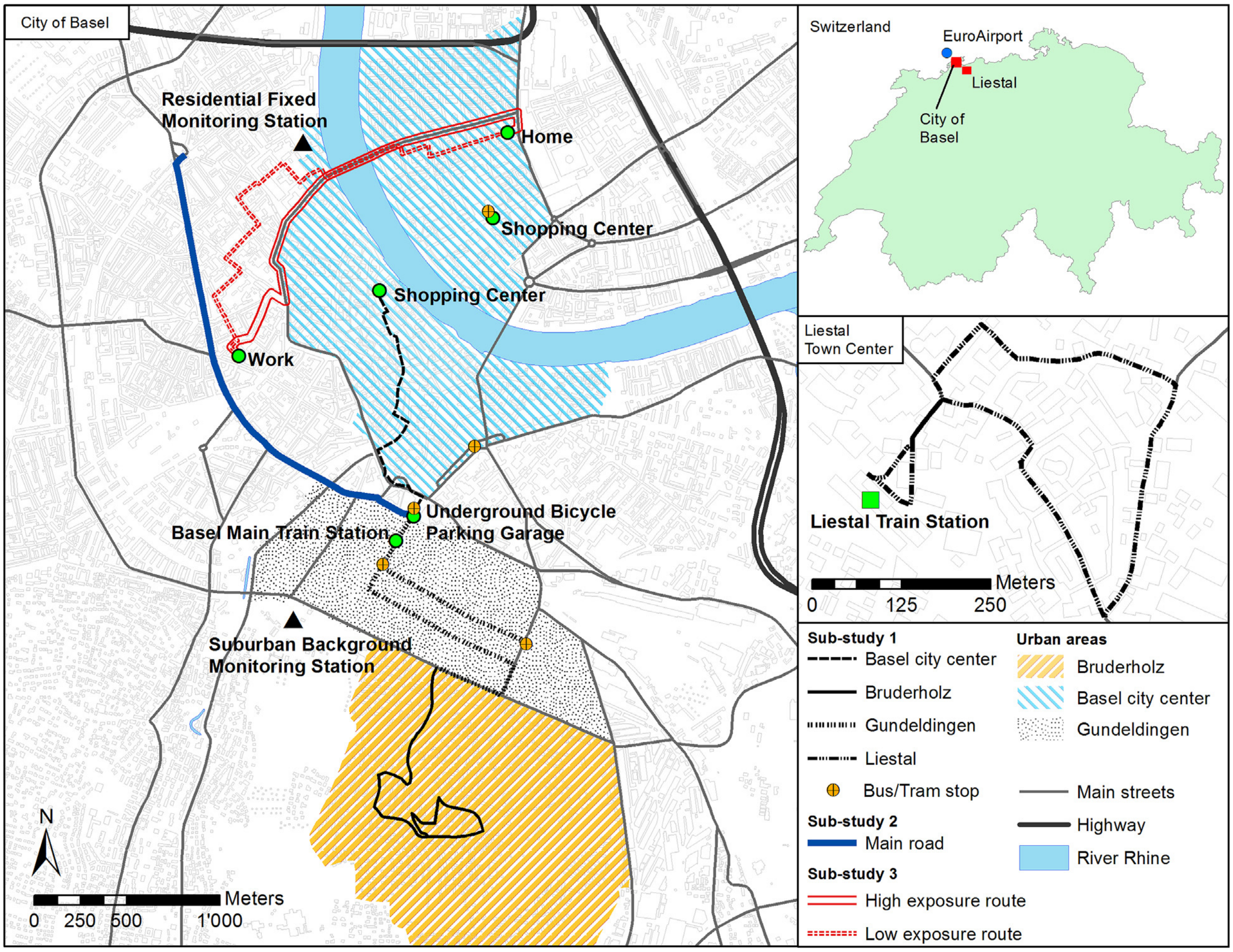

Fig. 1. Study area, urban areas, sampling routes and locations by measurement approach. A TSI CPC 3775 was used at the suburban background station and a TSI CPC 3022 at the residential fixed monitoring station.

ventilation and windows were open occasionally. Cars were gasoline-fueled Renault Modus (models 2008 and 2010), with windows closed, air conditioning off and the ventilation system on a moderate level.

2.4. Sub-study 3: 24-h personal UFP measurements

A total of twenty-four 24-h personal measurement campaigns were carried out by one person. Measurements were equally distributed over weekdays (Monday–Friday) across three seasons – winter, spring and summer. Two different bicycle commuter routes were defined to examine how routes influence personal exposure during commutes and its contribution to total daily exposure. During each sampling day, the person traveled twice along a potentially *high* (primarily main roads) and a *low* (avoiding main roads) exposure route along urban streets between home and work place during rush hours. A measurement day included one trip from home to work and back in the morning, and an additional trip to work in the morning with the return in the evening. In each season, during the first week, the trips from home to work followed the *low* exposure route while the trips back to home were done on the *high* exposure route. During the second week these routes were switched.

2.5. Exposure measurements and instrumentation

A newly developed portable device, the miniature Diffusion Size Classifier (miniDiSC), was used to measure total particle number concentration and average particle size distribution diameter in the size range of 10–300 nm with a sampling interval of one second. The device has been shown to agree within 20% ($R^2 = 0.90$) with standard condensation particle counters (TSI model 3775) (Fierz et al., 2011). The customized miniDiSCs used in this study had a battery life of 36 h. Inlets were placed near the breathing zone, on a shoulder strap of a backpack or on the headrest of the front passenger seat in the car (more details on instrument handling and inlets in Appendix B). Throughout the measurement campaigns, UFP concentration along with weather data were also collected from the city's suburban background station (condensation particle counter (CPC) 3775, TSI Inc., MN, 10-min averages) and another fixed station in a residential area (CPC 3022, TSI Inc., MN, 30-min averages) (Fig. 1). A summary of ambient UFP concentrations and meteorological conditions during each sub-study is provided in Appendix C.

2.6. Data analyses

Data were checked for unreliable measurements and outliers, using the miniDiSC software and information from the time-activity diaries filled out during measurements. Data cleaning steps and information used from the time-activity diaries are described in Appendix D.

One-minute averages were calculated for both UFP concentration and average particle size. The statistical analysis was then performed based on median UFP concentrations and average particle size by location (sub-study 1) and commute (sub-study 2). Results are presented in the text as average median UFP concentration (±standard deviation). For sub-study 3, both average and median UFP concentrations were computed for diurnal profiles (30-min), commutes, and the non-commuting time of each 24-h sample.

Since measurements were made at slightly different time points in different microenvironments and outdoor locations in the *first sub-study*, they were adjusted for intradiurnal temporal variation by computing their ratio to the simultaneously measured UFP at the suburban background station and multiplying this ratio with the daily median UFP concentration at the suburban background station (more details on ratios in Appendix E).

For *sub-study 2*, multivariate models with a random effect for sampling date were built to adjust for potential differences in weather conditions between sampling days and to correct for the fact that measurements were not taken simultaneously for the five transport modes. UFP concentration was log-transformed and regressed against time of the day/week (categorical variable), travel mode and residential fixed UFP levels (corresponding to the same measurement window) (general model). Temporal variables were selected based on the Akaike information criterion. Then, stratified models for each temporal category were computed after testing interactions of mode of transport and temporal variables using the likelihood ratio test.

In *sub-study 3*, the contribution of bicycle commuting to total daily (24 h) exposure was calculated separately for the *low* and *high* exposure commuter routes by the following equation:

$$C_{\mathrm{tot}}T_{24\ \mathrm{h}} = C_{\mathrm{mc}}T_{\mathrm{mc}} + C_{\mathrm{ec}}T_{\mathrm{ec}} + C_{\mathrm{nc}}T_{\mathrm{nc}},$$

where $C_{\mathrm{tot}}T_{24\ \mathrm{h}}$ is total daily cumulative exposure, C_{mc} and C_{ec} are the median morning and evening commute exposures along the same route between home and work, respectively, and C_{nc} is the exposure during non-commuting time periods (i.e. 24-h minus commute time). T_{mc}, T_{ec} and T_{nc} are the time windows for morning, evening commute and non-commuting periods, respectively. Two same weekday measurements within a season were combined to have morning and evening trips for both routes. The non-commute UFP exposure, however, was computed using the medians of both days.

The statistical software STATA (version 10.1) was used to perform the statistical analyses. Microsoft Excel 2003, Sigmaplot 11.0 and ESRI ArcGIS 9.3 were used in addition to produce the figures.

3. Results

3.1. Spatial variation of UFP by urban area and commuter microenvironment

The final dataset included 17 sampling days. The distribution of adjusted median UFP concentration and average particle size by outdoor sidewalks and other commuter microenvironments is provided in Table 1. The overall adjusted average median outdoor UFP concentration during walking in four different urban areas in the region of Basel was 14,143 ± 7725 particles cm^{-3}. A lower mean UFP concentration (12,609 particles cm^{-3}) was observed at the suburban background station for the same sampling period except in the *residential green* area, *Bruderholz*. Average median UFP concentration at *Bruderholz* was 43% lower than in the *urban residential* area *Gundeldingen* at the foot of *Bruderholz* hill. UFP variability was largest during walking in the *Liestal town center (traffic)* and in the *urban residential* area *Gundeldingen*. Large UFP variability was also seen at the bus and tram stops. Unadjusted UFP levels were highest at the train station and underground bicycle parking garage but were not affected directly by vehicular traffic.

In general, mean particle size distribution diameters showed an inverse relationship with UFP concentrations and were similar in all outdoor areas, except *Bruderholz (residential green)*, where the distribution shifted towards larger particles (Table 1). Among all areas *Liestal town center (traffic)*, had the smallest particle sizes.

3.2. Five modes of transportation

In total, data were collected from 275 valid trips. The average trip duration for walking, cycling, bus, tram and car was 31.8, 10.4, 7.4, 10.8 and 7.0 min, respectively. Boxplots of UFP concentration and particle size by transport mode and sampling period are shown in Figs. 2 and 3. Overall, median trip UFP ranged from 4188 to 161,942 particles cm^{-3}. Personal UFP levels were highest during car rides (31,784 ± 25,255 particles cm^{-3}), and lowest during bus rides (14,055 ± 7951 particles cm^{-3}). Personal average UFP levels were similar while walking (19,481 ± 11,705 particles cm^{-3}) and in the tram (18,818 ± 8120 particles cm^{-3}). In comparison, mean UFP concentration at the residential fixed monitoring site over the entire sampling duration was 14,420 ± 5536 particles cm^{-3}. For all transportation modes, the exposure contrast was highest during rush hour and lowest during weekend.

Multivariate regression models showed both transportation modes and sampling times as significant determinants of personal

Table 1
Ultrafine particle number concentration and average particle size during four outdoor walks and in commuter microenvironments.

	n (days)	n (minutes)	UFP concentration (particles cm^{-3})				Average particle size (nm)			
			Mean (sd)	Median	Min	Max	Mean (sd)	Median	Min	Max
Outdoor walks[a]										
Bruderholz, *residential green*	8	241	9′469 (5′377)	10′178	2′613	19′152	54.9 (10.4)	52.4	45.5	81.2
Basel city center, *pedestrian*	8	150	13′906 (4′625)	15″32	6′454	18′914	46.9 (7.2)	46.1	37.7	59.9
Gundeldingen, *residential urban*	8	227	16′481 (8’286)	15′702	6′019	32′328	47.8 (7.2)	46.1	35.8	61.5
Liestal town center, *traffic*	9	165	18′508 (8′960)	13′725	8′554	33′609	43.3 (6.4)	46.0	31.9	49.6
Suburban background[b]	17	1′818	12′609 (8′161)	11′177	1′506	37′144				
Microenvironments										
Bus/Tram stop (outdoor[a])	16	207	17′173 (8′724)	15′282	4′634	39′342	46.4 (8.6)	45.4	31.7	62.5
Train station (semi-outdoor)	17	238	25′629 (11′580)	23′993	8′197	58′092	49.8 (6.6)	49.7	37.1	65.9
Shopping centers (indoor)	7	210	7′672 (3′964)	7′063	2′229	17′181	50.4 (7.8)	49.2	40.3	70.0
Airport (indoor)	9	227	14′315 (7′691)	10′534	8′203	31′954	44.6 (11.0)	45.4	30.5	67.0
Bicycle parking garage	15	153	19′005 (6′109)	18′196	11′989	35′388	47.6 (9.4)	46.3	33.9	71.5

sd: standard deviation.
[a] Outdoor measurements have been adjusted by the ratio of daily median UFP concentrations and simultaneously measured UFP concentrations at the suburban background station.
[b] Simultaneously measured UFP concentration measured at the suburban background with a TSI CPC 3775.

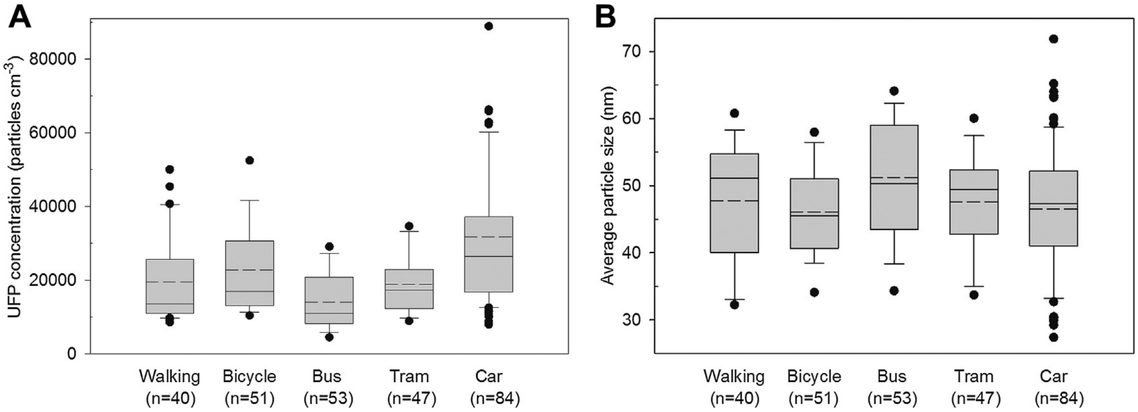

Fig. 2. (A) Ultrafine particle number concentrations and (B) average particle size by transportation mode for entire sampling period. Boxplots are based on the distribution of the median ultrafine particle number concentration and average particle size measured across all trips. Boxes represent the 25th to 75th percentile, the dashed central line the mean, whiskers represent the 10th and 90th percentiles. The points are outliers outside the 10th and 90th percentiles.

UFP levels during commute. Adjusted geometric mean (GM) ratios between each of the four transport modes and walking, as well as adjusted GM ratios between weekday and weekend sampling times are shown in Table 2. In the general model, adjusted GM ratios were highest for car or bicycle depending on time period and lowest during bus rides. The UFP levels in all transport modes were highest during morning rush hour, followed by evening rush hour and non-rush hour – all on weekdays – and the lowest on weekends. In the models stratified by sampling period, the smallest GM ratio was found for bus travel (bus:walking = 0.58) during the morning weekday rush hour.

Average particle size in the five transportation modes ranged between 46.1 and 51.2 nm (Fig. 2). Average UFP particle diameters followed an opposite trend than UFP concentration, showing larger average particle sizes for transportation modes and sampling times with lower UFP number concentrations and vice-versa. During weekday rush hour, especially in the morning, smaller average particle sizes were measured for bicycle (42.6 ± 6.5 nm) and walking (43.0 ± 7.9 nm) trips compared to other travel modes.

3.3. 24-h personal measurements

Overall average 24-h mean (7988 ± 3071 particles cm^{-3}) and median (5780 ± 2859 particles cm^{-3}) personal UFP exposures were lower than the average daily levels at the outdoor residential fixed station (11,556 ± 2608 particles cm^{-3}). Average median UFP concentrations along the *high* (primarily main road) and *low* (avoid main road) exposure routes were 34,025 ± 26,406 particles cm^{-3} and 18,156 ± 8615 particles cm^{-3}, respectively. Moreover, commute exposure levels were higher in winter than spring and summer, whereas non-commute personal levels were lower in the winter than in other seasons (Appendix Table F). Average trip duration for the *high* exposure route was 13 and 11 min in the morning and afternoon, respectively. In comparison, the *low* exposure route was longer by two minutes in either direction.

Overall, average particle size of these personal measurements was 63.5 ± 16.8 nm, which corresponds to the particle sizes measured during non-commuting times (Appendix Table F). Average particle sizes during commutes were 46.1 ± 10.9 nm and 49.0 ± 9.2 nm, for the *high* and *low* exposure routes, respectively.

Fig. 4 displays 30-min mean and median personal UFP levels and simultaneously measured mean outdoor UFP concentrations at the fixed residential monitoring station. Out of the four commutes per sampling day, only the first morning commute to work and the last evening trip to home were included in the analysis to represent a typical weekday commute. The excluded commutes were replaced by seasonal indoor UFP levels at home and place of work. Personal UFP levels followed the ambient diurnal pattern but were ~50% lower. Peaks in personal UFP levels were related to morning and evening commutes. Indoor activities such as cooking and candle burning may have further elevated UFP concentrations in the evening. During morning commutes, the personal mean UFP (*high* exposure route) was similar to the fixed station, yet exceeded ambient UFP by ~70% during the evening commute. Median values showed no difference between the two commuter routes. However, time-weighted median commuter exposures, computed using the

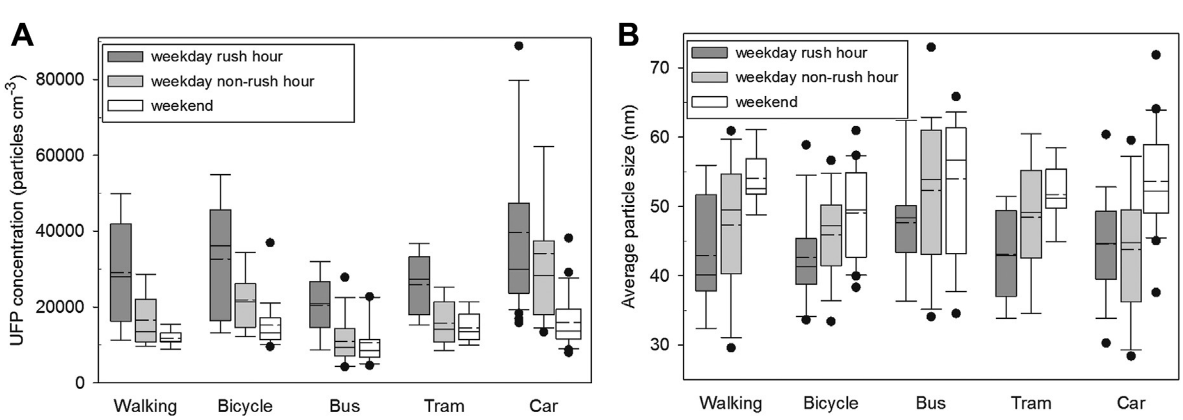

Fig. 3. (A) Ultrafine particle number concentrations and (B) average particle size by mode and sampling periods. (Figure description same as Fig. 2.)

Table 2
Multivariate regression models for ultrafine particle number concentration (particles cm^{-3}) with random effect for sampling day for total sampling time (general model), and stratified by four sampling periods (geometric mean GM (95% Confidence Interval)).

	General model	Weekday rush hour AM[a]	Weekday rush hour PM[b]	Weekday non rush hour	Weekend
Nr of trips	275	53	44	96	82
Nr of sampling days	18	11	10	12	6
Independent variables					
Residential fixed station[c]	1.41 (1.05–1.90)*	1.55 (1.37–1.76)*	1.65 (1.18–2.31)*	1.42 (1.01–2.01)*	1.56 (1.31–1.85)*
Travel mode					
Walking (reference)	1	1	1	1	1
Bicycle	1.23 (1.07–1.42)*	1.30 (0.98–1.74)	1.26 (0.86–1.83)	1.21 (0.93–1.57)	1.24 (0.99–1.54)
Bus	0.71 (0.62–0.82)*	0.58 (0.45–0.74)*	0.90 (0.61–1.33)	0.65 (0.51–0.83)*	0.82 (0.64–1.04)
Tram	1.01 (0.87–1.17)	0.75 (0.57–0.98)*	1.10 (0.75–1.63)	0.98 (0.76–1.26)	1.18 (0.93–1.51)
Car	1.23 (1.04–1.47)*	1.05 (0.75–1.47)	1.48 (1.03–2.13)*	1.68 (1.20–2.36)*	1.22 (0.97–1.53)
Time of day/week					
Weekend (reference)	1				
Weekday non rush hour	1.24 (1.01–1.51)*				
Weekday PM[b] rush hour	1.39 (1.12–1.73)*				
Weekday AM[a] rush hour	1.86 (1.47–2.35)*				
Intercept (10^{-3})	14.1 (11.6–17.1)*	29.0 (21.0–40.2)*	17.2 (13.0–22.9)*	16.2 (12.6–20.7)*	13.1 (10.5–16.2)*

The likelihood ratio test between the general model and the general model with interactions between modes and time of day/week variables was significant ($p = 0.0189$). Significant differences between transport mode for all four sampling times were confirmed by WALD tests (Weekday rush hour AM $p = 0.0000$; weekday rush hour PM $p = 0.0487$; weekday non rush hour $p = 0.0000$; weekend $p = 0.0006$).
*p-value < 0.05.
[a] AM = morning.
[b] PM = afternoon.
[c] UFP concentration data from the fixed site was centered by the mean and log2 transformed.

actual trip durations, showed differences between the routes (Fig. 5). Cycling along main streets between home and work place ($\sim$24 min day^{-1}) contributed 21% to total daily exposure in winter and 5% in summer. Avoiding main roads ($\sim$28 min day^{-1}) reduced the contribution of bicycle commutes to total daily UFP exposures by $\sim$50%.

4. Discussion

This study provides an overview of personal UFP exposures near roads in different urban areas, transport microenvironments, modes of transport and street types during different times of the day, week and year in Basel. Sidewalk UFP concentrations were higher in the densely populated urban residential area and in the town center of Basel metropolitan area than in the green residential area with less traffic density. Higher UFP concentrations were observed for car travel and cycling compared to walking and public transportation. Levels were highest for all travel modes during morning weekday rush hours. Among all transportation modes, the bus showed the lowest UFP concentrations during all time periods. UFP exposure during bicycle commutes contributed to more than 20% of the total daily exposure in winter and could be reduced by

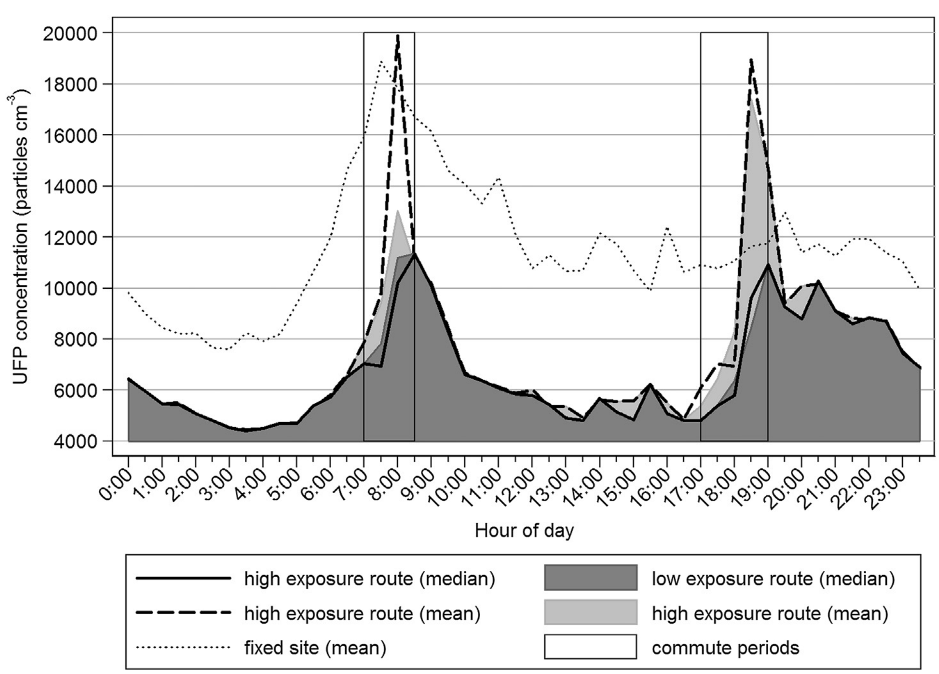

Fig. 4. Diurnal profile of 30-min median and mean ultrafine particle number concentration for bicycle commute along the high and low exposure routes and 30-min average particle number at fixed site.

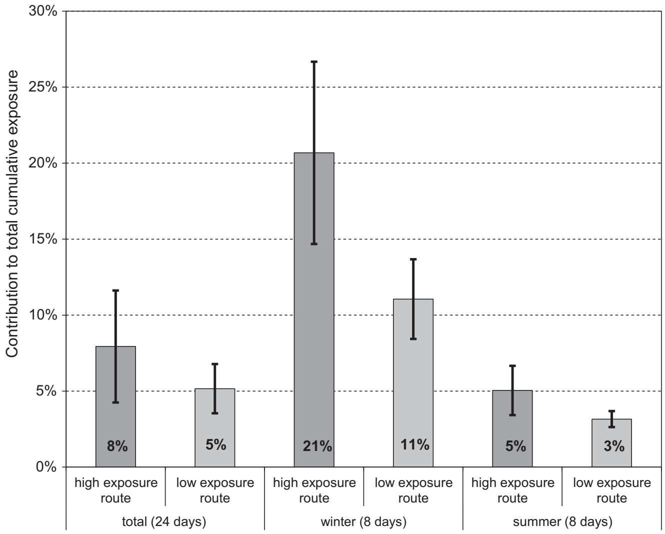

Fig. 5. Contribution of commute to total daily ultrafine particle number exposure. Error bars represent 95% confidence intervals derived using bootstrap with 100 replications.

avoiding main roads. In general, smaller average particle sizes and high UFP levels were measured at places and for travel modes with proximate traffic.

4.1. Spatial variation in urban areas and microenvironments

Only a few studies have examined mobile sidewalk exposures across different urban areas (Boarnet et al., 2011; Buonanno et al., 2011). Previously described factors affecting near-road UFP levels, such as vehicle density, composition, and building structure (Boarnet et al., 2011; Buonanno et al., 2011), seem to explain UFP variability among the four urban areas in this study.

Elevated indoor UFP concentrations in the (non-smoking) airport, compared to the shopping centers, could be due to infiltration of ambient UFP concentrations through natural and mechanical ventilation systems. Particles originating from flight activities and ground support vehicles could also be potential contributors. Westerdahl et al. (2008) also documented increased UFP levels in the 10–20 nm particle size range near airports.

4.2. Comparison between transportation modes

UFP concentrations in car and/or on bicycle in this study were similar to reported levels in Brussels, Belgium (Int Panis et al., 2010), the Netherlands (Boogaard et al., 2009; Zuurbier et al., 2010) or Denmark (Vinzents et al., 2005), and more than 60% lower than in Barcelona (de Nazelle et al., 2012) or London (Kaur et al., 2005). This is not surprising, as the latter two cities are characterized by higher traffic density and a higher proportion of (heavy duty) diesel vehicles than Basel.

Over the entire monitoring period, adjusted UFP levels were similar for bicycle and car travel (for both, ratio to walking was 1.23). Insignificant differences between bicycle and car UFP levels were also reported in two of three Belgian cities (Int Panis et al., 2010). Other studies that used a car with similar ventilation settings found slightly higher UFP concentrations in car compared to bicycle (Kaur et al., 2005; Knibbs et al., 2011). Briggs et al. (2008), however, reported lower UFP levels in car than during walking, in London.

In addition to the mode of transport, the time of day and day of the week significantly affected UFP concentrations, which is due to well-characterized diurnal patterns (Morawska et al., 2008) and traffic volume (Knibbs et al., 2011). During morning rush hours, the bicycle:walking ratio (1.30) was slightly higher than the car:walking ratio (1.05), but the opposite was true for weekday afternoon rush hours and non rush hours. Temperature is known to affect UFP concentrations through condensation or evaporation of semi-volatile compounds, with a stronger association observed for cycling than for automobiles (Knibbs et al., 2011). Cooler temperatures during the early morning rush hours may explain the higher bicycle:walking ratio. Furthermore, cyclists and pedestrians are exposed to cooler temperatures than those traveling by car, likely contributing to this observation.

The lowest UFP levels were measured for bus travel, in contrast to other studies (Kaur et al., 2005; Knibbs and de Dear, 2010). UFP concentrations in bus were especially reduced during morning rush hours, compared to walking. The elevated in-bus UFP concentrations observed in previous studies were primarily explained by the self-pollution of diesel-powered buses without particulate filters (Knibbs et al., 2011). However, the Basel bus fleet is composed of mechanically ventilated CNG buses – the bus type with the lowest UFP concentrations (Knibbs et al., 2011) –, and diesel buses fitted with particulate filters. UFP levels were higher in trams than in buses, probably due to different ventilation characteristics (partly open windows), more stops, and the trams' position on the middle of the road. UFP levels in tram were similar to walking, but showed less variability. Only during morning rush hour when windows were closed did the tram have significantly lower UFP concentration than walking. Newer trams with improved air conditioning and filtration may decrease a passenger's UFP exposure.

Similar to urban sidewalk measurements, UFP level had an inverse association with particle size, which remained consistent regardless of transportation mode or time of day. Few studies have quantified particle size diameters in different transportation modes. Buonanno et al. (2011) reported a mode diameter of 30–40 nm for pedestrians in Cassino, Italy; in this study, the average particle size was 48 nm. In comparison, for other modes of transport – bus, tram and car – particle sizes were either similar or higher. Larger particle sizes were observed for bus travel, likely due to less infiltration of smaller particles in the well encapsulated vehicle cabins (Knibbs et al., 2011; Zhu et al., 2007).

The comparability of UFP levels among studies might be impaired by the different measurement devices used, primarily due to different particle cut-off diameters. So far, P-Traks and CPCs 3007 are the most commonly used particle number counters in personal in-transit studies. In this study, however, a miniDiSC was used, which is a good alternative to handheld CPCs; it neither requires a working liquid nor a horizontal orientation. Moreover, it is lighter and has >24 h battery life versus 6–8 h for CPCs. The miniDiSC has been shown to be more accurate than P-Trak, especially for particles <40 nm (Meier et al., 2013). The miniDiSC comparison to CPC 3007 showed good agreement ($R^2 \sim 0.99$), with average differences between −8 and +25%. Differences were most pronounced for low concentrations (Asbach et al., 2012).

4.3. Contribution to total daily exposure

To our knowledge, no other study has quantified the contribution of commute exposures to total daily personal UFP exposures by 24-h measurements. A direct comparison of the measurements to the residential fixed station monitor, however, underestimates the personal UFP levels because of the higher cut-off diameter of the device used at the fixed station. By scaling the two devices, the personal commute mean UFP levels will be even more elevated than those captured by the residential fixed monitor. Higher mean UFP levels were observed for bicycle commutes along the *high* exposure route than for those along the *low* exposure route. This was not true when expressed in 30-min median concentrations. While measuring mean exposures during a limited time frame may be representative of the total exposure for that time window, it cannot be guaranteed that they are representative of the general commuting situation (other roads, other time windows). For instance, the influence of one or a few unusually heavy emitters passing by while monitoring cannot be ruled out. Most in-transit studies report mean concentrations, which may be very specific to the commutes and individuals. Median UFP concentrations were also computed in this study due to the non-normal data distribution and to ensure the representativeness of the UFP exposures for the general commuting situations.

4.4. Further exposure measures

The time spent in a certain transport mode is an important determinant of commuter exposure. Taking travel time into account, the time-weighted total commute exposures were the highest for walking but similar for car, tram and bicycle travel (Appendix Table G). Another proxy for assessing personal commuter exposure is the inhaled dose of UFP. To use the data in a health risk assessment context, one may apply dose-oriented approaches as, for example, in de Nazelle et al. (2012). Factors determining the biologically relevant dose of particle deposition in the respiratory tract depend (besides travel duration and concentration levels) also on physiological factors such as breathing rate and tidal volume, thus, on the physical effort (Hofmann, 2011). Previous research has reported 2–4.5 times higher respiratory minute ventilation in cyclists compared to motorized transport commuters, resulting in a higher UFP dose per distance traveled for cyclists than for car and bus occupants (Int Panis et al., 2010; Zuurbier et al., 2010). Physical activity also modifies the deposition pattern of particles, favoring the lower airways, which is also governed by decreasing particle size (Hofmann, 2011). Active commuters may therefore have a higher particle deposition of the smaller particle fraction of UFP in the alveolar region. Thus, alveolar dose of the smallest UFP might be further amplified during cycling compared to more sedentary travel modes (bus, tram). Consequently, the health benefit of choosing routes with low UFP concentrations may be even larger than what the exposure data may indicate.

4.5. Strengths and limitations

This study has several strengths. First, to our knowledge it is the first study to provide comprehensive information on average particle sizes in various microenvironments within urban areas. Second, it is the only study that compares UFP levels and average particle sizes for five commuter modes on the same route. Finally, it is the first study to carry out 24-h personal UFP monitoring to evaluate the contribution of commute exposures to total daily personal exposures for different bicycle routes.

A limitation of this study is the rather small sample size and sampling duration, especially for sub-study 1, even though samples included all seasons, different times of day and days of the week. Furthermore, measurements in the urban areas, for different modes and routes were neither carried out simultaneously nor randomly for practical reasons. This may have induced systematic differences in transportation mode and urban area measurements because of temporal factors. While UFP concentrations for both sub-study 1 and sub-study 2 were corrected for these temporal fluctuations, micrometeorological differences due to wind direction and speed could not be taken into account. An additional limitation of the study was having only one subject for all the 24-h personal measurements (sub-study 3), which makes it difficult to estimate UFP exposure or inhalation dose for the whole population. However, the study does provide important insights into commuter UFP exposure and its implications for total daily UFP exposures. Additional and larger studies are warranted to further validate these findings.

5. Conclusions

As in most other studies, UFP exposure has been shown to be higher during daily commutes compared to other daily activities and suburban background concentrations. The average particle size is generally smaller in microenvironments with proximate traffic, which also supports its relevance for health. In addition, commute exposure depends on the time spent in a certain mode of transport, urban area and time of day and week. Exposure of cyclists can be reduced when avoiding main roads, especially during elevated ambient UFP concentrations in winter and during morning rush hour.

While there is evidence of adverse health effects of short-term UFP exposure, long-term population effects of both in-transit and total daily exposures are not well understood (Hoek et al., 2010). To assess the long-term effect of UFP exposure, detailed information on commuter behavior (travel mode, time of the day, route, waiting time at public transport stops, duration) together with detailed data on UFP concentrations and time spent at various home and work indoor environments is essential. Furthermore, due to the spatial variability of UFP concentrations within the city, monitoring stations in different urban areas with different road and traffic characteristics (vehicle types, volume) are recommended to accurately assess population exposure.

Acknowledgments

We are thankful to Mark Davey from University of Washington and Alex Ineichen and Chantal Wullimann from Swiss TPH for their help in field work. This work is part of the European wide project Transportation Air pollution and Physical ActivitieS: an integrated health risk assessment progamme of climate change and urban policies (TAPAS), which is funded by the Coca-Cola Foundation. We also acknowledge the Federal Office of Environment (BAFU), Switzerland for partial funding and supplying fixed site data.

Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2013.05.003.

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4 MODELING OF LONG-TERM COMMUTER EXPOSURE TO TRAFFIC-RELATED AIR POLLUTION

Article 2: Simulation of NO_2 population commuter exposure to NO_2 using different air pollution models

This paper has been published:

Ragettli M. S., Tsai M.-Y., Braun-Fahrländer Ch., de Nazelle A., Schindler Ch., Ducret-Stich R. E., Ineichen A., Perez L., Künzli N., Phuleria H. C., 2014. Simulation of population-based commuter exposure to NO_2 using different air pollution models. International Journal of Environmental Research and Public Health, 11(5), 5049-5068.

Int. J. Environ. Res. Public Health **2014**, *11*, 5049-5068; doi:10.3390/ijerph110505049

OPEN ACCESS

International Journal of
Environmental Research and Public Health
ISSN 1660-4601
www.mdpi.com/journal/ijerph

Article

Simulation of Population-Based Commuter Exposure to NO_2 Using Different Air Pollution Models

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Received: 11 February2014; in revised form: 12 April 2014 / Accepted: 29 April 2014 / Published: 12 May 2014

Abstract: We simulated commuter routes and long-term exposure to traffic-related air pollution during commute in a representative population sample in Basel (Switzerland), and evaluated three air pollution models with different spatial resolution for estimating commute exposures to nitrogen dioxide (NO_2) as a marker of long-term exposure to traffic-related air pollution. Our approach includes spatially and temporally resolved data on actual commuter routes, travel modes and three air pollution models. Annual mean NO_2 commuter exposures were similar between models. However, we found more within-city and within-subject variability in annual mean (±SD) NO_2 commuter exposure with a high resolution dispersion model (40 ± 7 µg m^{-3}, range: 21–61) than with a dispersion model with a lower resolution (39 ± 5 μg m^{-3}; range: 24–51), and a land use regression model (41 ± 5 μg m^{-3}; range: 24–54). Highest median cumulative exposures were calculated along motorized transport and bicycle routes, and the lowest for walking. For estimating commuter exposure within a city and being interested also in small-scale variability between roads, a model with a high resolution is recommended. For larger scale epidemiological health assessment studies, models with a coarser spatial resolution are likely sufficient, especially when study areas include suburban and rural areas.

Keywords: air pollution; model comparison; traffic; travel mode; travel pattern

1. Introduction

Daily travel within urban areas is an important component of human exposure to traffic-related air pollutants. Levels of traffic-related air pollutants such as nitrogen dioxide (NO_2), ultrafine particles (UFP), and carbon monoxide (CO) have consistently been shown to be higher in urban areas and in transit-related environments than at other non-occupational locations [1–4]. In Europe, people spend about 8% of the day in transport environments [5]. Many of those daily trips, especially the commutes to and from work or school, generally take place during times of the day with peak traffic flow and thus high concentration levels. In most epidemiological studies on health effects of long-term exposure to traffic-related air pollution, however, in-transit exposure is ignored [6]. The exposure assessment of these studies mostly relies on estimated levels at no more than one fixed-site per person such as average level of the respective pollutant at the person's home or a fixed monitoring station nearby, or on traffic indicator variables, including distance to major roads and traffic intensity [6]. With technological advances and the development of personal monitors, several personal monitoring studies have been carried out to better quantify air pollution exposures in traffic (e.g., [1,7–11]). Although personal exposure studies provide important insights into exposure determinants, such studies are generally not feasible for large cohort studies due to the high costs and the commitment required of the study participants.

Only a few modeling approaches exist to estimate air pollution exposure in transit. Some long-term exposure assessment studies have applied the concept of microenvironments to take into account in-transit exposure (so-called compartment models). This approach uses the average concentrations within different transport environments, derived from personal or fixed station measurements, and multiplies them by the time spent in such microenvironments. While some studies differentiate between several transportation modes [12], others use only a general "transport" microenvironment [5]. For both approaches, uncertainties remain for pollutants with high spatial and temporal variability within microenvironments, such as for example NO_2 concentrations, thus creating inter-subject variability [13,14]. More dynamic models account for people's specific location throughout the day along with time-activity information and in-transit patterns. Exposures are estimated by overlaying air pollution models with information from census data, time-activity and/or geo-coded origin-destination information from surveys [13,15–17]. Another approach integrates activity-based transport models simulating spatially and temporally resolved vehicle volume, traffic emissions, and population density to predict population exposure [18–20]. Limitations of such exposure simulations include the imprecision of spatial in-transit data. Some models simulate trips as straight lines or the shortest or fastest route along roads between locations and zones without knowing the exact route and/or travel mode. Others are based on a synthetic population and routes are generated stochastically as in the case of activity-based transport models.

Simulated in-transit exposure estimates might be further impaired by the limited spatial resolution of the air pollutant models used which do not accurately represent the high spatial variability of pollutant concentrations, especially in urban streets [18,21]. Developing models of high resolution requires expertise, adequate data and can be costly. Air pollution models commonly used to assess long-term exposure to traffic-related NO_2 include inverse-distance weighted interpolation of monitoring data (e.g., [15]), dispersion models (e.g., [17,22]) and land use regression (LUR) models (e.g., [23,24]). While dispersion models mostly rely on dispersion theory, emission and meteorological data, LUR models apply regression techniques using actual air pollution monitoring data and predictor variables obtained from geographic information systems (GIS). More recently, hybrid models were also developed (e.g., [25]) which combine personal or regional monitoring with other air pollution modeling methods [26].

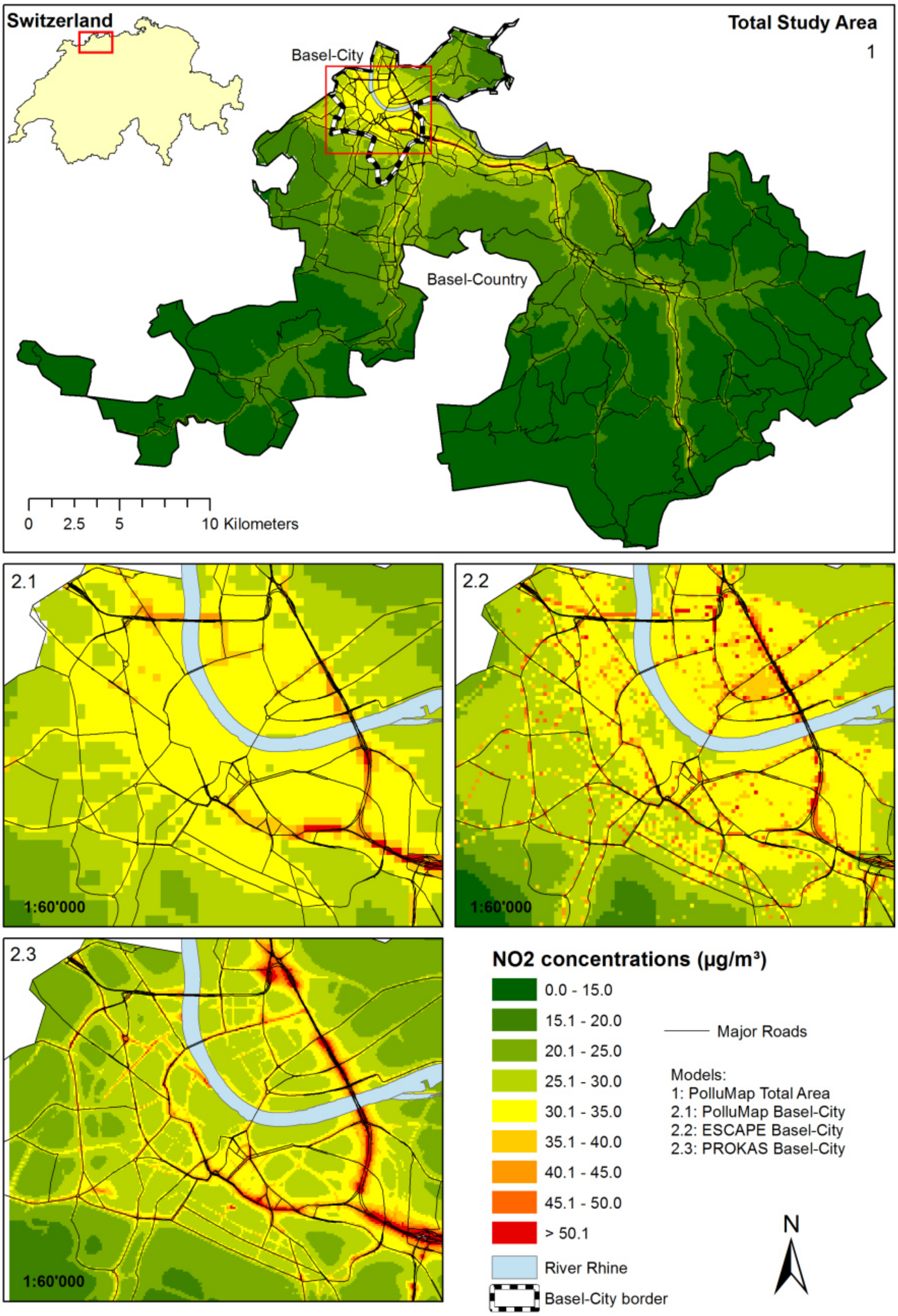
Better quantification of daily in-traffic exposure of a general population is important to provide better estimates of total air pollution exposures in investigations of long-term health effects. The aim of this study was to develop an approach to estimate individual NO_2 exposures in a representative sample of the population during commute within the metropolitan area of Basel (Switzerland). Our approach includes spatially and temporally resolved data on commuter trips within the study area, and three annual air pollution models with varying spatial resolution. This paper describes the simulation of commuter routes and the in-transit NO_2 exposure from outdoor origin. It also evaluates the differences between these NO_2 commuter estimates for the three models that may occur when applying them in long-term exposure assessments. The potential bias that can occur when ignoring these commute exposures but rely on home outdoor locations only in epidemiological studies on the long-term health effects of traffic-related outdoor air pollution is explored in Ragettli *et al.* [27]. As in many epidemiological studies, we chose NO_2 as marker for traffic-related outdoor air pollution as it describes the spatial distribution of traffic-related air pollution well. But, in principle, the simulation is applicable to any other traffic-related pollutant.

2. Methods

2.1. Study Area

Our study was carried out in the region of Basel (Switzerland), which covers the two Swiss Counties (called Cantons) of Basel-City and Basel-Country (Figure 1). The area (550 km^2) includes a population of 465,000 people. While the Canton of Basel-City is a predominantly urban area with buildings of usually three to five stories, Basel-Country is largely suburban and rural in character. Hereafter, we differentiate Basel-City from the total study area and present results separately. The region is a relatively low-pollution area with an annual mean NO_2 suburban background concentration of 23.5 $\mu g\ m^{-3}$ in 2010.

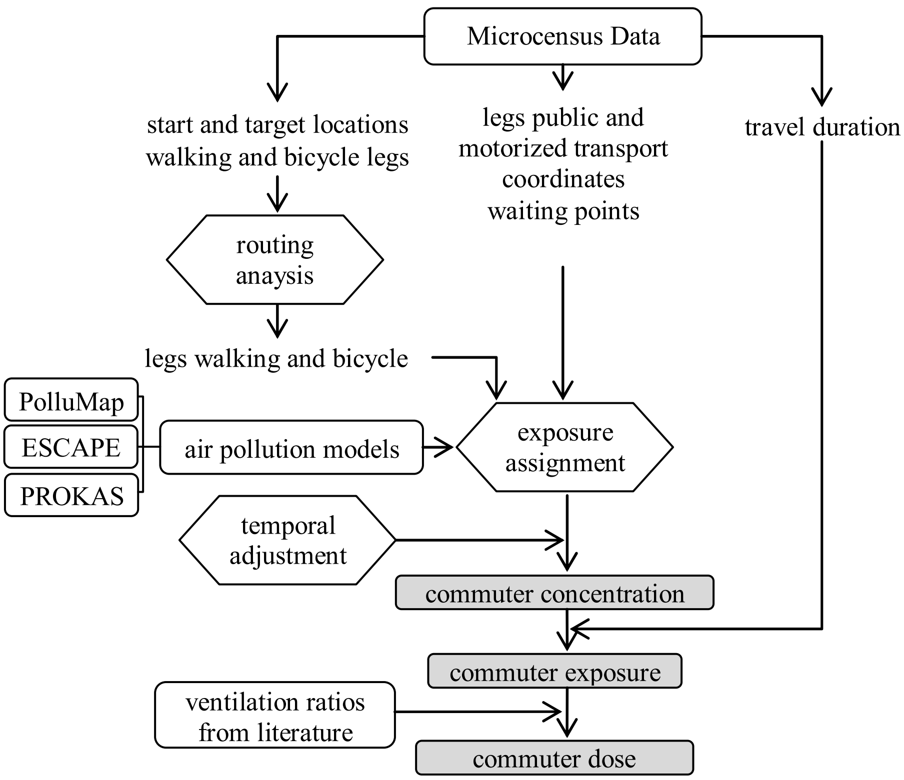
Figure 1. Annual mean NO_2 concentrations from different air pollution models for total study area (**1**); and Basel-City (**2**).



2.1.1. Study Population and Commuter Routes

The study methodology is illustrated in Figure 2. We extracted information on commuter routes from the year 2010 Swiss Mobility and Transport Microcensus [28]. Our focus was on commutes between home, work or school locations as those trips account for a large fraction of travel time on work days and are usually carried out regularly over time. The telephone-based survey included geo-coded time-activity diaries covering one day of a representative number of randomly selected individuals of each Swiss Canton. Geo-codes were recorded for start locations, trip destinations and places where study participants changed their mode of transport during trips. In addition, the actual route of public transport and motorized transport legs were simulated based on the coordinates by an interactive routing tool during the interview. A leg is defined as each contiguous part of a trip that is covered with the same travel mode. For example, a trip of a person who walks to the train station, takes the train and then walks to work from the destination train station covers three legs. The routing was performed based on the TeleAtlas MultiNet road network and a public transport network with integrated time table. All public transport and motorized transport routes ≥3 km were verified during the interview.

Figure 2. Schematic representation of the applied methodology. Boxes are inputs, and hexagons are analysis steps. Shadowed boxes indicate commuter estimates.



We subsequently simulated both walking and bicycle routes based on the geo-coded start and target locations using the GIS based route finder Network Analyst by ESRI (ArcGIS 9.3, Redland, CA, USA). The routing was performed using the Swiss GIS road network VECTOR25 (Federal Office of Topography swisstopo, Wabern, Switzerland, 2008) which has been shown to be more complete than the TeleAltas road network for smaller side streets and pedestrian roads [29]. The shortest routes between the geo-codes were determined using the distance, *i.e.*, the road segment length, as the cost factor in the analysis. We validated the routing performance of the GIS model by comparing real commutes of test persons with the simulated routes (for details, see Supplementary Section 1). Additionally, a quality check of the simulated distance *versus* reported distance of the Microcensus data of all legs and travel modes assured that large detours were avoided. Comparisons between the reported distance and routing distance of walking and bicycle legs showed moderate to high agreement (R^2 = 0.6 to 0.8) for both the microcensus data and validation study (see Supplementary Section 2).

To evaluate the benefit of verifying car legs ≥3 km instead of modeling the shortest routes in terms of driving time between origin and destination location, we also simulated the fastest route based on the TeleAtlas road network for subjects who travel only by car between home and work/school locations (results of this sub-analysis are provided in the Supplementary Section 3).

We classified each commuter leg as either a main or a side street based on the longest road segments of the underlying road network. Major streets in TeleAtlas were defined as the functional road classes (FRC) 0–4. In VECTOR25, streets classified as highways and class 1 roads were used as main roads. Public transport legs that were not directly overlapping with the TeleAtlas road network were classified based on the length of the nearest road segments within a buffer of 15 m.

A total of 736 subjects (28% of all respondents with time-activity information in the study area) were selected based on the following inclusion criteria: (a) living and working, or attending a school within the study area, locations not being the same place; (b) reporting at least one trip between home and work location within the study area classified by purpose working or education; (c) quality of geo-codes being of sufficient quality (house number or street level). Indirect commuter trips, including for example a stop at a shop or day care, were also included. If an uneven number of trips between home and work/school location were reported (11% of the total), *i.e.*, a trip from either home to work or vice-versa was missing, the reported single trip was duplicated. For these cases the time of the day when leaving home, work or school was used as time information.

2.2. Air Pollution Models

We used three spatially resolved annual mean ambient air pollution models to estimate exposure to NO_2 during commute. The models were all originally developed for estimating outdoor air pollution exposure at home outdoor locations. Two models for Basel-City and one for the total study area were available (see Table 1). The first model, PROKAS, was developed for the calculation of traffic induced air pollution for the Basel department of air hygiene. It consist of a Gaussian plume model (PROKAS_V) to estimate the urban traffic background concentration for a given road network and meteorology, and an integrated building structure module (PROKAS_B) [30]. The latter is used to account for the rather complex built environment of urban areas. It is based on pre-calculated dimensionless concentrations for 20 different building structures and 36 air flow directions determined by the microscale dispersion model MISKAM [31]. Additional NO_2 concentrations such as household (heating), shipping traffic from the river Rhine, industry and commerce were estimated with the three-dimensional model LASAT [32] and overlaid with the traffic-related NO_2 concentrations. The road transport emissions for all major roads were computed by a local traffic model (mobility department Basel-City) and projected to the TeleAtlas road network. The second model, ESCAPE, was developed within the framework of the European Study of Cohorts for Air Pollution Effects (ESCAPE) using LUR modeling based on 2009 NO_2 measurement data at 40 locations [33]. Given that the model was designed to estimate NO_2 exposures at home outdoor locations and not in transport environments *per se*, we applied the LUR model to a 50 × 50 m grid corresponding to the quality of the model input data. More information on the Basel ESCAPE model is provided in the Supplementary Section 4. Finally, a nationwide dispersion model, PolluMap, was available from the Swiss Federal Office for the Environment (FOEN). The nationwide model computes source-specific annual concentrations based on a Gaussian plume model using emission inventories from 2010, a national road network map and meteorological data. Emission inventories considered include road traffic, rail traffic, aviation, industry, commerce, construction, household (heating), agriculture and forestry [34].

Table 1. Characteristics of the three air pollution models used to individually assign commute exposure.

	Models		
	PROKAS	**ESCAPE**	**PolluMap**
Year	2010	2009	2010
Grid size	25 × 25 m	50 × 50 m	100 × 100 m
Method	Gaussian dispersion, integrated building characteristics	Land use regression	Gaussian dispersion
Availability	Basel-City	Basel-City	Switzerland
Comparison with measurements	NA	R^2 = 0.67 [a]	R^2 = 0.80 [b]
Reference	Air Hygiene Department Basel and Lohmeyer 2008 [30]	Beelen *et al.* 2013 [33]	Federal Office for the Environment Switzerland (FOEN) [34]

Note: [a] unadjusted R^2; [b] Measured values are the arithmetic mean of the three annual averages 2008, 2009, 2010.

2.3. NO_2 Exposure Assessment

We overlaid maps (Figure 1) of annually averaged ambient NO_2 concentrations from the three air pollution models on the commuter legs to estimate commuter exposure. NO_2 concentration of a leg (C_{leg}) was computed based on the sum of the extracted NO_2 grid concentrations (C_{grid}) weighted by the length of the leg within the grid (Equation (1)):

$$C_{leg} = \frac{1}{total\_length} \sum_{grid=1}^{m} C_{grid} \times length_{grid} \quad (1)$$

We calculated temporal adjustment factors for each hour of the day separately for main roads and side streets to consider the diurnal pattern of NO_2 levels and road-type specific differences in hourly traffic volume and composition of vehicles. NO_2 data (30-min averages) from two fixed air pollution monitoring stations, a street site and an urban background site within the Canton of Basel-City, were used to derive the ratios. Ratios were computed between the annual weekday hourly means and the annual mean concentration measured at the monitoring stations for main streets ($ratio_{m\text{-}h}$) and side streets ($ratio_{s\text{-}h}$) (for more details on ratios, street class distribution by travel mode see Supplementary

Section 5). We then applied ratios to each leg concentration C_{leg} based on the road classification and start hour of the leg to compute subjects' commuter NO_2 concentration, $C_{subject}$ (Equation (2)) and exposure, $E_{subject}$ (Equation (3)). For the calculation of subjects' commute exposure, waiting time between two legs (e.g., when transferring from one mode to another for example at public transport stops) and respective NO_2 concentrations (C_{wait}) were also considered:

$$C_{subject} = \frac{1}{n}\sum_{leg=1}^{n} C_{leg} \times ratio_{m,s-h} \tag{2}$$

$$E_{subject} = \sum_{leg=1}^{n} C_{leg} \times ratio_{m,s-h} \times t_{leg} + \sum_{wait=1}^{n} C_{wait} \times ratio_{m,s-h} \times t_{wait} \tag{3}$$

where t_{leg} is the duration spent on the leg, and t_{wait} the time spent at a waiting location. Time-weighted commuter exposure is defined as the exposure divided by the total commuter duration of a subject. We used the reported travel time and waiting time information from the microcensus data for all travel modes.

Finally, as a proxy for the inhaled dose, we derived adjusted estimates of exposure, taking into account mode-specific ventilation rates (we use the term "dose" hereafter). Since neither physical activity measures nor adequate data on body weight and body height were available, we applied ventilation ratios extracted from the literature to each leg. A ratio of 1.7 [8] for walking and 2.0 for bicycle [8,35], respectively, relative to public transport and motorized transport was assumed.

Comparisons between the three air pollution models based on subjects' commuter NO_2 estimates (*i.e.*, concentration, exposure and dose) and by travel mode (*i.e.*, legs without waiting time) were then performed to evaluate the potential differences in outdoor NO_2 estimates that may arise when applying models with varying modeling techniques, spatial resolution and input data. A validation of the in-transit NO_2 exposure estimates—for the overall population and by travel mode—was neither the purpose of this study nor possible due to the unavailability of reliable real-time personal NO_2 monitoring devices with appropriate sensitivity and specificity. As our focus was the long-term exposure to outdoor air pollution in transport environments, the benefit of validating the annual models with short-term personal measurements is limited. However, to evaluate the performance of the air pollution models, we compared the PROKAS, ESCAPE and PolluMap model to NO_2 measurements from a total of 31 monitoring sites within Basel-City from the Swiss study on Air Pollution and Lung and Heart Diseases in Adults (SAPALDIA) (see Supplementary Section 6). These measurements were conducted outside subjects' homes in three biweekly integrated sampling campaigns in 2011 using Passam passive diffusion samplers (Passam AG, Schellenstrasse, Männedorf, Switzerland). We compared the average ambient NO_2 concentrations of each site to the respective grid value of the three models. The data analyses were conducted using the statistical software STATA (version 12.1, STATA Corp., College Station, TX, USA).

3. Results

3.1. Commuter Behavior of the Study Population

The majority (84%) of the study population reported two commuter trips per day. The remaining population traveled four times per day between home and work/school locations. The average number of legs (±standard deviation (SD)) per subject and day in Basel-City and the total area was 4.6 (±3.0) and 4.6 (±2.9), respectively. A summary of the characteristics of the study population (age, sex, working hours per week) is shown in Table S5 in the supplement. In the total study area, the main travel modes used for the daily commute to work/school (defined as the mode used for the longest distance of the commute trips per day) were motorized transport (car and motorcycle; 32%) and public transport (bus, tram, train; 30%). However, within Basel-City, the active transport (walking: 27%, bicycle: 30%) was the main travel mode, followed by public transport (32%). Motorized transport was used by 9% of the subjects living and working in Basel-City.

The average daily commuting distance within Basel-City was about half of that of the total study area (Table 2). However, the average trip duration between home and work/school locations (18.2 ± 11.5 min in Basel city) was only 14% shorter. Average daily travel time for all main travel modes were rather similar within Basel-City (30–35 min), except for public transport, which was about twice as long (62 min). Commuting mainly took place within the rush hours 6–8 am and 4–6 pm (Figure S4), coinciding with the diurnal peaks of air pollution.

Table 2. Daily commuter distance and commuter duration of subjects per main travel mode and study area.

	Basel-City					**Total Area**				
	n (subjects)	**mean**	**(sd)**	**min**	**max**	**n (subjects)**	**mean**	**(sd)**	**min**	**max**
commute distance (in m)										
all modes	258	6,086	(4,588)	52	29,095	736	13,976	(15,329)	23	88,346
walking	69	2,965	(2,239)	328	16,126	140	2,480	(2,043)	23	16,126
bicycle	78	5,325	(3,583)	52	26,426	131	5,627	(3,910)	52	26,426
motorized transport	22	9,128	(4,128)	3,569	17,136	234	21,318	(17,610)	877	88,346
public transport	83	8,801	(5,082)	3,261	29,095	219	19,081	(15,204)	2033	83,182
other	6	3,153	(1,981)	1,316	6,310	12	2,882	(2,259)	1061	7,895
Commute duration (in minutes)										
all modes	258	42	(25)	4	155	736	49	(33)	2	204
walking	69	35	(24)	9	155	140	32	(25)	2	155
bicycle	78	30	(15)	4	90	131	32	(19)	4	125
motorized transport	22	35	(14)	19	64	234	43	(26)	4	163
public transport	83	62	(24)	23	140	219	78	(32)	23	204
other	6	32	(17)	20	63	12	31	(20)	6	74

Note: sd: standard deviation; min: minimum; max: maximum.

3.2. Comparison of Air Pollution Models

In the overall comparison between the model-based NO_2 estimates and the SAPALDIA NO_2 measurements, the PROKAS model obtained best agreement (R^2 = 0.58) whereas correlations were lower but similar for the ESCAPE (R^2 = 0.41) and the PolluMap model (R^2 = 0.46). While the PROKAS model predicted the street sites concentrations better than the other models, the urban background sites showed good agreement also with the nation-wide dispersion model PolluMap, which had the lowest resolution (see Supplementary Section 6).

Summary statistics of estimated time-weighted subjects' commuter NO_2 exposure during commute using the three air pollution models are shown in Table 3. Within Basel-City, mean and median NO_2 concentrations and exposures were similar between the models. However, as illustrated by the standard deviations and confirmed by the Fisher Pitman test, the variability and range of model estimates were significantly increased with higher model resolution. Covering the total study area, the PolluMap model also allowed comparisons of within Basel-City commuter exposures to commutes within the total study area, *i.e.*, including subjects traveling between the two Cantons and within Basel-Country. Average exposure estimates from the PolluMap model for the total study area were ~5 µg m^{-3} lower than within Basel-City, and the range was twice as large because of the smaller values on the lower end.

Table 3. Summary of time-weighted subjects' NO_2 exposure during commute (in µg m^{-3}) for Basel-City by air pollution model, and for the total area (only one model available).

	Model	n (subjects)	mean	(sd)	min	p5	median	p95	max
Basel-City	PROKAS	258	39.9	(6.5)	20.7	29.3	40.1	49.7	61.4
	ESCAPE	258	40.8	(5.4)	23.8	31.8	41.3	49.7	53.8
	PolluMap	258	38.8	(4.7)	24.1	30.3	39.2	46.0	51.0
Total area	PolluMap	736	33.7	(7.7)	12.4	19.8	34.8	45.0	52.2

Note: sd: standard deviation; min: minimum; p5: 5th percentile; p95: 95th percentile; max: maximum.

In general, both leg and subject specific NO_2 concentrations correlated well between the models (r = 0.81–0.91, Table S6). NO_2 concentrations from PROKAS showed higher correlations with ESCAPE (the second highest resolution model) than with PolluMap, the lowest resolution model. Spearman correlation coefficients of subjects' NO_2 commuter exposures and dose estimates were almost identical for all model pairs and were close to 1.0 (Table S6).

As illustrated in Figures 3 and 4, we observed a non-linear relationship between model estimates. Compared to PROKAS, a systematic underestimation of subjects' highest NO_2 commuter estimates and overestimations of the lowest values in both PolluMap and ESCAPE models was found. The relationship of the NO_2 commuter concentrations between the model pairs PolluMap-PROKAS was best fitted by a quadratic function (R^2 = 0.70), and between ESCAPE-PROKAS by a cubic function (R^2 = 0.77). Average differences (and SD) between time-weighted NO_2 commuter exposure estimates of the three model pairs PolluMap-PROKAS, ESCAPE-PROKAS and PolluMap-ESCAPE within Basel-City were 0.97 (±3.12), −1.08 (±3.71) and −2.04 (±2.61) µg m^{-3}, respectively (Figure 4). Differences were significantly different from 0 (tested by a Wilcoxon signed rank test).

Figure 3. Scatter plot comparing subjects' estimated commuter NO_2 concentration based on the high spatial resolution model (PROKAS) with the estimates from PolluMap and ESCAPE models, respectively, using subjects from Basel-City (*n* = 258).

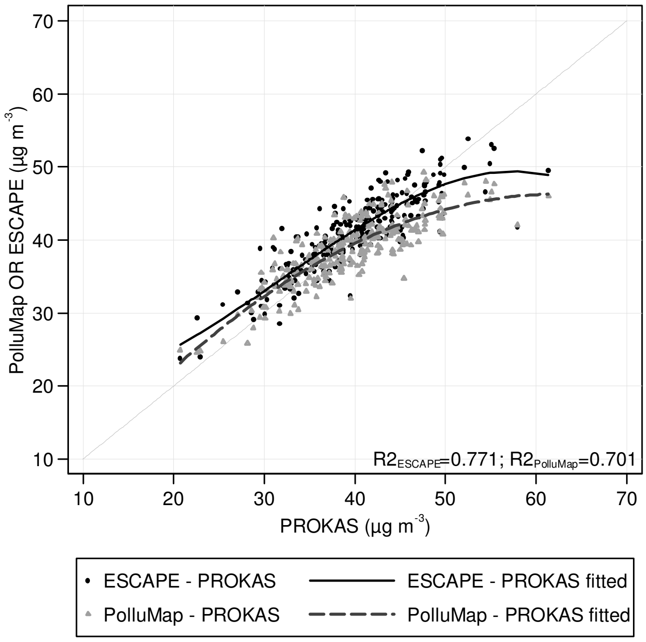
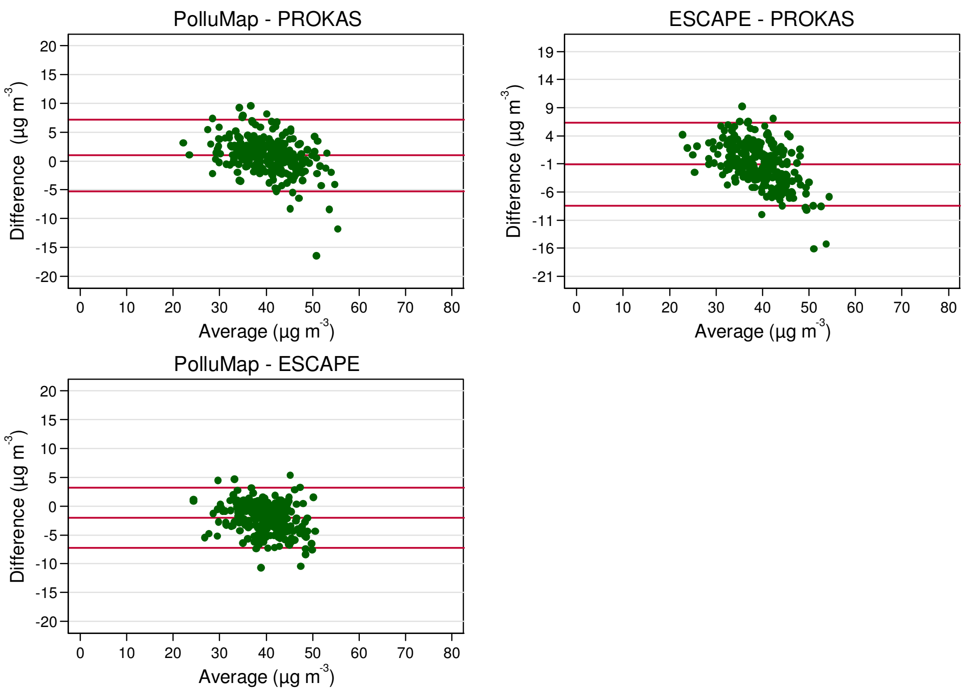


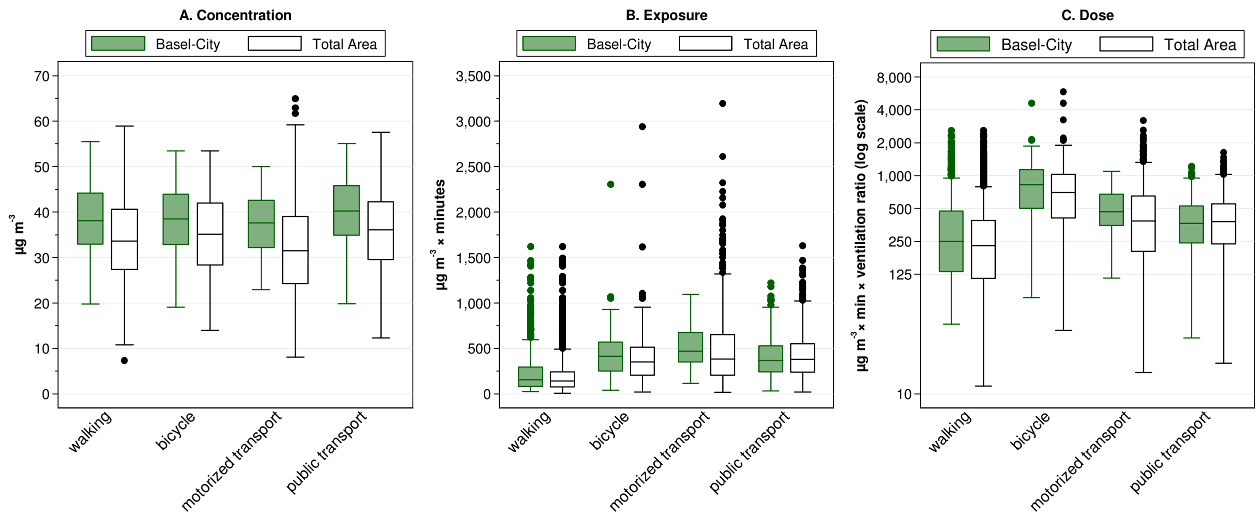
Figure 4. Bland Altman plots of time-weighted commuter NO_2 exposure of subjects commuting within Basel-City (*n* = 258). The lines represent the mean difference ±2 × standard deviation.



3.3. Commuter NO_2 Concentration, Exposure and Dose by Travel Mode

The number of legs within Basel-City (and total study area) by travel mode walking, bicycle, motorized transport and public transport were 636 (1,614), 204 (385), 58 (602), and 259 (735), respectively. Based on the commuter legs, in-transit concentration, exposure and dose are displayed by travel mode in Figure 5. Results are shown for the PolluMap model to allow comparisons between study areas. Within Basel-City, the median NO_2 commuter concentrations estimated by the PolluMap model for walking, bicycle and motorized transport were rather similar (~38 μg m^{-3}), and were slightly lower than for public transport (40 μg m^{-3}). A different modal pattern emerged when considering the travel time spent in the travel modes. Highest median cumulative exposures (*i.e.*, the concentration multiplied by the duration) with the PolluMap model were obtained for motorized transport (468 μg m^{-3} × minutes) and bicycle (414 μg m^{-3} × minutes), and the lowest for walking (156 μg m^{-3} × minutes). The highest median dose was observed for bicycle commutes (829 μg m^{-3} × minutes × ventilation ratio) in the model where a two-fold increase in minute ventilation was assumed for bicycle *versus* public and motorized transport. Walking remained the mode with the smallest dose (266 μg m^{-3} × minutes × ventilation ratio), although a ventilation ratio of 1.7 relative to motorized transportation was applied. In the total study area, the modal pattern was similar to the one in Basel-City, albeit mode-specific NO_2 estimates were generally lower.

Figure 5. Box plots of in-traffic NO_2 concentration (**A**); exposure (**B**); and dose (**C**) by travel mode and study area using the PolluMap model. Estimates are based on commute legs: boxes represent 25th to 75th percentile, central line the median, bars outside the box represent the most extreme values within 1.5 × the inter quartile range of the nearer quartile, and circles are outliers.



With the higher resolution model, PROKAS, more variability in travel mode-specific commuter NO_2 estimates was observed (data not shown). In addition, for the active transport legs—more often happening on side streets—the PROKAS model obtained 1%–2% lower estimates than the PolluMap model. In contrast, PROKAS provided 5%–6% higher estimates for passive transport legs which happen more frequently on busy roads. The percentage of the legs assigned as road class main roads within Basel-City to walking, bicycle, motorized and public transport legs was 23%, 35%, 52% and 61%, respectively (Table S2).

4. Discussion

The exposure to traffic-related air pollution during commute of the population living and working within Basel-City and Basel-Country was estimated using spatially and temporally resolved commuter route data, information on travel modes used and three NO_2 air pollution models with different spatial resolutions. Within Basel-City, estimated average time-weighted population exposure was similar between all models (around 39–41 µg m^{-3}). Compared to the dispersion model with the highest resolution, both the LUR model (applied to a 50 × 50 m grid) and the nation-wide dispersion model PolluMap (grid size 100 m), underestimated the concentrations on the higher end, and overestimated the values on the lower end. In the total study area, including also Basel-Country, average time-weighted commuter exposure estimated with just the PolluMap model was 34 µg m^{-3}. Commuter estimates from the same model showed greater variability and covered a wider range in the total study area (12.4–52.2 µg m^{-3}) than within Basel-City (range: 24.1–51.0 µg m^{-3}).

Only a few studies have estimated NO_2 in-transit exposures based on travel routes. De Nazelle *et al.* [36] extracted NO_2 exposures from an annual dispersion model in Barcelona based on Global Positioning System (GPS) tracks from 36 working adults. The temporally adjusted in-transit exposure was twice as high as our estimates within Basel-City, illustrating both higher in-transit NO_2 concentrations and the higher urban background NO_2 concentration level in Barcelona (Spain). In Flanders and Brussels (Belgium), Dhondt *et al.* [19] predicted an average in-traffic population exposure of 38 µg m^{-3} over the total area using an activity-based transport model. In an exposure simulation study at census tracts level in Vancouver (BC, Canada) , annual average hourly means of NO_2 levels were 34 µg m^{-3} on highways and arterial roads and 26 µg m^{-3} on less important roads using a dispersion model and census data [15].

To our knowledge, this is the first time that three air pollution models with different spatial scales were compared for estimating commuter exposure in the same area. We found more within-city and within-subject variability in NO_2 concentrations with the city-specific dispersion model PROKAS than with the LUR model and the nation-wide PolluMap dispersion model. LUR models have been shown to better reflect the spatial variability of traffic-related pollutants within an urban area than conventional dispersion models [25,37] or inverse-distance weighted interpolation of monitoring data [15,23]. Compared to dispersion models, less spatially resolved input variables are required for LUR models to accurately predict within-city variability of traffic-related NO_2 [26,37]. In our case, the NO_2 PROKAS dispersion model performed somewhat better in our commuter exposure simulations. Beelen *et al.* [33] showed that the accuracy of LUR models to predict NO_2 concentrations depends on the quality of the monitoring data and/or GIS variables. In particular, local traffic-intensity data have been shown to be important for achieving good model performance. The moderate model R^2 of 0.67 of the LUR model used in this study is likely reflected by the limited availability of traffic input variables and possible the limited contrasts in traffic density in the City of Basel. The comparison with measurements from street sites supports this finding. However, it must be emphasized that, our LUR model was applied at a grid resolution of 50 × 50 m; therefore, the decrease in model performance may be due to both, the chosen resolution and the intrinsic limitation of the LUR model. In addition, our validation of the models with fixed-site NO_2 measurements is not fully appropriate for the typical exposure during commute because measurements do not represent the concentrations on the traffic routes but rather home outdoor concentrations.

Comparing the two dispersion models, the model with the higher resolution showed greater variability between commuter exposures, and better agreement with measurements at street sites. The inclusion of traffic data and meteorological parameters at a more local scale and additional consideration of the building structure likely explain the higher variability, and wider range in commuter exposure estimates of the PROKAS model, and also the higher validity observed in the comparison with street sites measurements. Dispersion models have difficulties to predict within-city contrasts when interpolating meteorological data from sparse weather stations and from emission inventory data of low resolution [25]. Underestimation of NO_2 concentration at street sites was also observed earlier in the previous version of the PolluMap model (year 2000, 200 × 200 m) [22].

Our comparison of the three models in Basel based on simulated commute exposure estimates suggests that the decision on the model to be used to estimating commuter exposure in long-term epidemiological studies depends on the aim of the study and the size and geographic diversity of the study area. For estimating commuter exposure within urban areas and examining small-scale variability between road classes, a model with a high resolution representing well the urban street environment is recommended. This seems to be especially relevant for exposure assessments within a city, where inclusion of local traffic variables of sufficient quality (hourly traffic counts, street configurations) in the model is indispensable. For larger scale longitudinal epidemiological health assessment studies, however, models with a coarser spatial resolution might be sufficient, especially when a study area is comprised of a mix of urban, suburban and rural regions. Also, higher resolution dispersion models that include detailed traffic and 3D building data, are particularly costly to develop, need adequate expertise and are often limited in spatial coverage.

Our in-transit NO_2 estimates of long-term exposure were not validated with personal measurements. In line with the vast majority of epidemiological studies on long-term health effects of air pollution, our evaluation relies on the accuracy of the ambient models rather than personal measurements. Our objective was the estimation of exposure to air pollution during commute, using NO_2 as the marker of traffic-related air pollution. As in the epidemiological studies, we were not interested in total personal exposure to NO_2 *per se* as this would describe a mixture of exposure to pollution from traffic, gas cooking and other sources of combustion. Accordingly, our approach relies on the same ambient models used to derive home outdoor concentrations. Additional improvements in commuter exposure estimates may be expected when combining modelling methods with personal exposure data, as for example in hybrid models [26].

The strength of this study is the detailed data on travel behavior of a representative subset of the population. We had spatially and temporally resolved data on each leg of a commuter trip including information on travel modes used, time of day and locations where the mode of transport was changed. Our comparison of the cumulative NO_2 commuter exposures and doses by leg shows considerable differences between travel modes and thus indicates the importance of differentiating between travel modes and related routes and travel times. Furthermore, unlike other exposure simulation studies, the estimation of commuter exposure was based on real geo-coded travel routes of a population. In this study, motorized and public transport legs comply closely with actual travel routes and are not based on assumptions. Simplified trip simulations in other studies such as the shortest route or straight line between two locations, zones or census tracts [15–17] may add uncertainties as drivers may prefer other routes avoiding red lights and congestions. In a short validation study with test persons (data not shown), car routes between home and work locations often did not correspond to the shortest or fastest route within the city of Basel. Thus, verifying car routes >3 km likely helps to prevent misclassification of air pollution exposure (Supplementary Section 3). However, walking and bicycle legs were also based on the shortest routing algorithm in this study. Cyclists, especially, may choose to avoid main roads and thus may have longer commuter distances. Several studies have shown that travelling by bicycle along a greener route reduces both exposures [7,38] and dose [39]. Therefore, exposure levels may be overestimated when assuming shortest routes [11]. Our comparison of the reported travel distance against routing distance, however, aimed to control for large route discrepancies (see Supplementary Section 2).

Our exposure simulation—besides potential inaccuracies of the air pollution models *per se*—had some sources of uncertainties. Comparison between travel modes are based on the spatial location of the route, distances and durations. We did not take into account travel microenvironments such as in-vehicle exposure modification due to the potential use of ventilation systems or the commuter's position on the road. Therefore, we may have over- or underestimated in-vehicle NO_2 concentrations. To our knowledge, there is no extensive measurement campaign of NO_2 exposures between travel modes available, and literature on in-vehicle exposure modification of NO_2 is very rare. Short-term measurements by Harrison *et al.* [9] in London found higher levels in buses (39 $\mu g\ m^{-3}$) than in cars (25 $\mu g\ m^{-3}$) or trains (16 $\mu g\ m^{-3}$). A study by Chan and Chung [40] found significant differences in the indoor:outdoor (I/O) ratio for various ventilation modes and outdoor environments when driving in Hong Kong. On urban streets, a mean NO_2 ratio of 0.8, 1.0 and 0.6 were reported for fresh-air intake, open windows, and air-recirculation, respectively. Ventilation characteristics of the vehicles vary by season and other vehicle characteristics. Therefore, an integration of different ventilation characteristics are expected to be small in the annual mean commuter estimates. In addition to ventilation characteristics, differences between mode-specific concentration levels and between studies vary by various factors such as meteorology, traffic parameters, and vehicle type, thus generalization from one study to another may not be appropriate [3]. A recent UFP monitoring study along a main road in Basel by Ragettli *et al.* [7] observed higher levels while driving a car or cycling compared to walking and public transportation. However, no consistent correlations between in-transit concentrations of UFP and NO_x have been reported [3], and therefore no modification was applied in this study. De Nazelle *et al.* [36] used ratios of BC concentrations between transportation microenvironments as a proxy for NO_2 ratios, which explains in part higher commuter exposures found in that study. Yet another limitation of this study was the relatively small area of the study. Commutes of the Basel population to other cities within Switzerland could not be included. It must be assumed that mean commute-related NO_2 exposure and dose would be higher when including people spending more time on their daily commutes especially when commuting on highways and in tunnels [12,21,41].

So far, epidemiological studies on long-term effects of ambient air pollution rely on home outdoor concentrations to estimate total exposure. The expansion of this approach to integrate outdoor concentrations at work or school addresses—the second most frequent location of time—is straightforward. Our approach targets at the improvement of total exposure estimates for epidemiological studies on long-term health effects through integration of the third most important time window, namely commute related exposure to ambient air pollutants. The average annual time-weighted commuter exposure estimates (34–41 μg m^{-3}) in the total study area were higher than the annual mean NO_2 concentration at the suburban background (24 μg m^{-3}) but similar to the urban background station in Basel-City (30 μg m^{-3}). Therefore, the contribution of commute to total NO_2 exposure and the related effect on long-term health outcomes might be small for the majority of the population in Basel. However, for some subgroups of the population the commuter exposure could be more important, as indicated by the range of NO_2 exposures (Table 3). Further studies may expand toward the integration of other microenvironments such as time activity patterns during leisure time.

5. Conclusions

We provide an approach to simulate commute routes and related exposure to traffic-related NO_2 that can be used to improve both in-transit exposure estimates and total daily exposure estimates for epidemiological studies assessing long-term effects of air pollution on health. Information to be collected from the study population should include home and work location, travel mode, travel behavior (number of trips within a day and week, travel duration) and route (fastest *versus* shortest route, detours, and habits on avoiding main roads). The relative contribution of these commuter estimates to total daily exposure needs to be investigated and further research is needed to validate such simulations.

The decision on which air pollution model to be used depends on the aim of the study, the local situation, and on practical issues. In general, it is important to gain an understanding of the available models, and to consider the type of information and uncertainty that could emerge when using one model over another. We recommend using air pollution models which represent well the urban street network within a city when being interested in small-scale variability and differences between travel modes. Our analysis indicates that for epidemiological health assessment studies over a larger geographic scale covering rural, suburban and urban areas, however, models with a coarser spatial resolution are likely adequate, but need to be formally evaluated.

Acknowledgments

This work is part of the European wide project Transportation Air pollution and Physical ActivitieS: an integrated health risk assessment program of climate change and urban policies (TAPAS), which is funded by the Coca-Cola Foundation. We also acknowledge the Federal Statistical Office for providing the microcensus data, the Air Hygiene Department Basel for supplying fixed site data and the PROKAS model, and the Federal Office of the Environment (FOEN) for providing the PolluMap model. NO_2 data were provided by Swiss National Science Foundation, grant number 135673 (SAPALDIA-Exposure assessment). We are also grateful to Jörg Jermann from the Civil Engineering Department Canton Basel-Country, Switzerland, for his GIS support.

Author Contributions

Martina S. Ragettli: data collection, NO_2 simulation, statistical analysis, manuscript writing; Ming-Yi Tsai: LUR modeling, collaboration manuscript writing; Charlotte Braun-Fahrländer:

local project coordinator; Audrey de Nazelle: TAPAS project coordinator, collaboration data preparation and analyses; Christian Schindler: statistical analysis support; Alex Ineichen: GIS support; Regina E. Ducret-Stich: collaboration to statistical analysis and manuscript writing; Laura Perez: collaboration data treatment and interpretation; Nicole Probst-Hensch: SAPALDIA Project Leader and study design; Nino Künzli: scientific supervisor and study design; Harish C. Phuleria: scientific supervisor and study design.

Conflicts of Interest

The authors declare they have no conflict of interests.

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Article 3: The relevance of commuter and work/school exposure in an epidemiological study on traffic-related air pollution

This paper has been published:

Ragettli M. S., Phuleria H. C., Tsai M.-Y., Schindler Ch., de Nazelle A., Ducret-Stich R. E., Ineichen A., Perez L., Braun-Fahrländer Ch., Probst-Hensch N., Künzli N., 2014. The relevance of commuter and work/school exposure in an epidemiological study on traffic-related air pollution. Journal of Exposure Science and Environmental Epidemiology, doi:10.1038/jes.2014.83.

Journal of Exposure Science and Environmental Epidemiology (2014), 1–8

www.nature.com/jes

ORIGINAL ARTICLE

The relevance of commuter and work/school exposure in an epidemiological study on traffic-related air pollution

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Exposure during transport and at non-residential locations is ignored in most epidemiological studies of traffic-related air pollution. We investigated the impact of separately estimating NO_2 long-term outdoor exposures at home, work/school, and while commuting on the association between this marker of exposure and potential health outcomes. We used spatially and temporally resolved commuter route data and model-based NO_2 estimates of a population sample in Basel, Switzerland, to assign individual NO_2-exposure estimates of increasing complexity, namely (1) home outdoor concentration; (2) time-weighted home and work/school concentrations; and (3) time-weighted concentration incorporating home, work/school and commute. On the basis of their covariance structure, we estimated the expectable relative differences in the regression slopes between a quantitative health outcome and our measures of individual NO_2 exposure using a standard measurement error model. The traditional use of home outdoor NO_2 alone indicated a 12% (95% CI: 11–14%) underestimation of related health effects as compared with integrating both home and work/school outdoor concentrations. Mean contribution of commuting to total weekly exposure was small (3.2%; range 0.1–13.5%). Thus, ignoring commute in the total population may not significantly underestimate health effects as compared with the model combining home and work/school. For individuals commuting between Basel-City and Basel-Country, ignoring commute may produce, however, a significant attenuation bias of 4% (95% CI: 4–5%). Our results illustrate the importance of including work/school locations in assessments of long-term exposures to traffic-related air pollutants such as NO_2. Information on individuals' commuting behavior may further improve exposure estimates, especially for subjects having lengthy commutes along major transportation routes.

Journal of Exposure Science and Environmental Epidemiology advance online publication, 10 December 2014; doi:10.1038/jes.2014.83

Keywords: air pollution; bias; inhalation dose; mobility; NO_2; transport

INTRODUCTION

In most epidemiological studies on health effects of long-term exposure to traffic-related air pollution, exposure during commuting and at non-residential locations is ignored.[1] The exposure assignment of these studies typically relies on residential neighborhood exposure according to home addresses, census tracts or postal codes. This choice is justified by the fact that a substantial amount of time is spent at home. In addition, the air pollution exposure of specific (and often most susceptible) groups of the population, such as the very young children, elderly people or spatially segregated groups, may be well represented by the residential area.[2] However, for more mobile population groups, such as working adults and school children, ignoring exposure to outdoor air pollution while away from home may lead to misclassification of exposure [3,4] and bias in health-effect estimates.[5] Using only air pollution exposure at home may in particular ignore potential hot spots of exposure to air pollution from outdoor sources that are encountered during daily activities, such as while at work or in school and during commute.[6–8]

Several studies have shown that traffic-related air pollutants, such as nitrogen dioxide (NO_2) or ultrafine particles, show high spatial and temporal variability.[3,9,10] People are indeed exposed to potentially high levels of those pollutants, especially in commuting environments.[11,12] Moreover, recent exposure assessment studies reveal that a significant proportion of the total inhaled dose of air pollution occurs during transport because of the increase in breathing rates during walking and cycling compared with more sedentary travel modes or activities.[3,13,14]

Attempts to more accurately quantify exposure by incorporating daily movements include spatio-temporal modeling[4,14,15] and personal monitoring.[6–8,16] Point-based location data provided in time-activity diaries provide the opportunity to link activity patterns to air pollution concentrations in models. Activity data, however, generally lack information on detailed travel routes between the activity locations. In addition, there is a trend towards real-time tracking of both exposures and activity patterns with portable measurement devices and global positioning system (GPS) receivers.[17] However, dynamic exposure assessment methods are rarely applied in large studies on the long-term effects of outdoor air pollution because of limitations related to cost, feasibility and participant burden.

The aim of this study was to assess the contribution of the commute (i.e., the time spent in traffic traveling between

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Received 20 May 2014; revised 19 August 2014; accepted 1 October 2014

home and work or school) and work/school location to the total long-term exposure to urban air pollution from outdoor sources, using NO_2 as the marker of exposure. Using spatially and temporally resolved commuter route and NO_2 concentration data from a census-based random population sample, we assessed the contributions separately according to subjects' main travel mode. In addition, we examined the potential bias in health-effect estimates that can occur in a population sample when outdoor pollution levels at home are used as estimates of total exposure, instead of also considering exposure while at work (or in school) and during commuting. We also explored the extent of these biases in models where ambient NO_2 concentrations were further adjusted during active commute (i.e., walking or cycling) to reflect the higher intake of air pollution while physically active. The study is restricted to the Basel area, which is one of the eight regions that has been participating for >20 years in the SAPALDIA study (Swiss Study on Air Pollution and Lung Disease in Adults), which has used home outdoor pollution modeling as the default.[18]

METHODS

Study Design

We separately estimated outdoor exposure to traffic-related NO_2 at home and work (or school) locations and during the commute for a representative population sample living and working (or attending school) in the region of Basel, Switzerland. The area (550 km^2) consists of two counties (called Cantons), Basel-City and Basel-Country, that constitute an urban-rural area with a population of 465,000 (Figure 1). We extracted information on commute routes, home, work and school locations from geo-coded 24-hour time-activity diaries from the 2010 Swiss Mobility and Transport Microcensus.[19] This national telephone-based survey includes coordinates of origin and destination locations, places where study participants changed mode of transport during trips, and geo-coded travel routes for a representative number of residents in our study area. For each trip, detailed information on travel modes, duration and hour of the day was available. By computing time-weighted NO_2 exposures, we explored how commute and the time spent at work/school affect NO_2 exposure estimates. As in many epidemiological studies, we use NO_2 as a marker of exposure to traffic-related air pollution of outdoor origin rather than NO_2 *per se*; thus, we are not considering NO_2 from other sources such as indoor smoking or cooking.

Commute Exposure Assessment

The NO_2 commuter exposure data was simulated in a previous study by Ragettli et al.[20] In brief, the data set includes a representative population sample of 736 subjects from the 2010 Swiss Mobility and Transport Microcensus survey[19] who live and work or attend a school within the study area (i.e., who commute within the region of Basel). For each subject, annual mean NO_2 concentration and exposure estimates for total trips and legs (i.e., contiguous parts of the trip with the same mode of transport) between home and work/school were estimated from the 24-hour time-activity diaries. Only trips between home and work/school locations were considered, as those trips are usually carried out regularly over time. Individuals' NO_2 exposures during commuting were computed by overlapping the geo-coded commuter legs with temporally adjusted estimates from the NO_2 annual mean map from the 2010 national Gaussian dispersion model PolluMap (100×100 m resolution).[21] The model has been successfully used in Swiss health research[18] and was found the best available model for estimating commuter exposure to traffic-related air pollution in both urban and rural areas in Basel.[20]

We extracted annual mean NO_2 concentrations for all outdoor locations at home, work and school from the same dispersion model at the corresponding geo-coded locations. Given our interest in the long-term contribution of commuting exposure to total urban air pollution exposure, we only included subjects working 50% or more outside of their homes, leaving 680 individuals for our analysis. A summary of the population characteristics is provided in Table 1.

Exposure Assignment

We assigned NO_2 exposure to each subject based on three figures: (1) outdoor concentration at the home address; (2) time-weighted home and

Table 1. Characteristics of the study population.

Characteristic	*Frequency/mean*
Total subjects (*n*)	680
Age (mean± SD)	36.1 ± 17.6
Female (%)	48.7
Living & working[a] within Basel-City (%)	35.3
Commuting between Basel-City and Basel-Country (%)	25.0
Living & working[a] within Basel-Country (%)	39.7
With two commuter trips/day[b] (%)	83.5
Working full time ($>=90\%$) (%)	58.2
Working 50–89% (%)	17.8
Student (%)	6.2
<15 years old (%)	17.8

[a]Or attending school. [b]The remaining subjects took four trips per day.

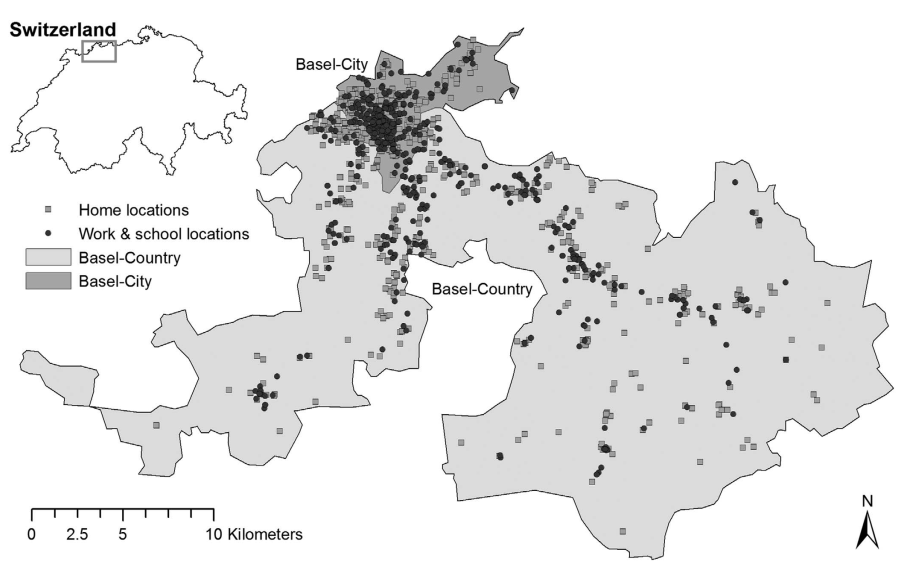

Figure 1. Study area with home and work/school locations. The total area consists of Basel-City and Basel-Country.

Table 2. Descriptions of the exposure ($_{exp}$) and inhalation dose ($_{dose}$) scenarios.

Scenario	*Simplified estimate*	*Assumed correct estimate*	*Description of estimates*
1	H_{exp}	HW_{exp}	H: home; HW: home, work/school
2	H_{exp}	HWC_{exp}	H: home; HWC: home, work/school, commute
3	H_{dose}[a]	$HWC_{dose\ moderate}$[b]	H: home; HWC: home, work/school, commute (moderate dose)
4	H_{dose}[a]	$HWC_{dose\ high}$[c]	H: home; HWC: home, work/school, commute (high dose)
5	HW_{exp}	HWC_{exp}	HW: home, work/school; HWC: home, work/school, commute
6	HW_{dose}[a]	$HWC_{dose\ moderate}$[b]	HW: home, work/school; HWC: home, work/school, commute (moderate dose)
7	HW_{dose}[a]	$HWC_{dose\ high}$[c]	HW: home, work/school; HWC: home, work/school, commute (high dose)

Example: in scenario 1, home exposure was compared with the time-weighted home and work/school estimates, which were assumed to be closer to the 'true' exposure. [a]Ventilation ratios applied: home location: 1; work location: 1 [b]Ventilation ratios applied: motorized transport: 1; public transport: 1; walking: 1.7; bicycle: 2; home and work location: 1. [c]Ventilation ratios applied: motorized transport: 1; public transport: 1; walking: 1.7; bicycle: 5.6; home and work location: 1.

work/school concentrations as a function of time spent at home and at work/school; and (3) time-weighted concentration that incorporates home, work/school and commute concentrations. Because information on commuting behavior was only available for 1 day of the year and information on workload was available on a weekly basis, we computed the estimates for a 7-day week. Hence, we calculated 5 days of commuting by replicating each participant's 1-day record to obtain a weekly estimate, assuming that work hours were evenly distributed over 5 days. For school children (age 6–14) and other students, we assumed that 6 h were spent at school per day. We assumed that the remaining time in seven 24-hour periods was spent at home.

Adjustment of Exposure During Physically Active Commute

Increased breathing frequency during cycling and walking results in higher intake of air pollution, and thus, ultimately increases the biologically relevant inhalation dose.[13] Therefore, for the time spent in active commute, we also derived adjusted estimates of exposure, now taken as a proxy for the dose (we use the term 'dose' hereafter). Our adjustment distinguishes two approaches, namely a *moderate* and *high* ventilation rate scenario for active commutes. Adjustment factors were derived from the literature on travel-mode-specific ventilation rates. For the *moderate* commuter dose estimates, minute ventilation (thus, NO_2 exposure) was assumed to be 1.7-fold higher while walking[11] and 2.0 times higher while on bicycle[11,22] than during commutes with motorized or public transportation, and time spent at home, work/school, which we considered as reference (no adjustment). For the *high* ventilation approach, we assumed a 5.6 times higher exposure on bicycle than the reference following the findings by Int Panis et al.[13] (derived by the mean plus standard deviation for males). For walking, we used the same ratio (1.7) than for the moderate ventilation rate scenario as we assume less variability in minute ventilation compared with cycling due to usually shorter distances.[20]

Contribution of the Commute

We explored the relative contribution of commuter exposure to total exposure—that is, the cumulative exposure considering home, work/school, and commute and the corresponding time spent in those microenvironments,—with and without the above described adjustment for inhalation rates over 1 week. We calculated the contribution separately according to the subjects' main travel mode, which was defined as the mode used for the greatest distance of commuter trips per day.

Scenarios

We calculated the bias in health-effect estimates that may occur when either outdoor NO_2 exposure at work/school or both NO_2 exposure at work/school and during commuting are ignored, using the seven scenarios described in Table 2. In scenario 1, we compared the traditionally used surrogate measure of outdoor exposure at home (H_{exp}) to exposure estimates that include both time spent at home and work/school (HW_{exp}), assuming that the latter is closer to the 'true' exposure. Similarly, we compared outdoor exposure at home (H_{exp}) with time-weighted home, work/school and commuter exposure (HWC_{exp}) (scenario 2). We also performed the same comparison (between residence-only estimates and estimates that include time spent at work/school and commuting time) for the dose estimates (i.e., cumulative exposures adjusted by ventilation rate), assuming a moderate commuter dose ($HWC_{dose\ moderate}$) and a high commuter dose ($HWC_{dose\ high}$) in scenarios 3 and 4, respectively. Finally, we computed the bias that occurs from using combined home and work/school exposure (HW_{exp}) and dose (HW_{dose}) compared with estimates that incorporate home, work/school and commuter behavior (HWC_{exp}, $HWC_{dose\ moderate}$, $HWC_{dose\ high}$) (scenarios 5, 6, 7).

Bias Factor Assessment

It is well known that associations between health outcomes and exposures may be estimated with bias if exposures are represented by surrogate measures.[5,23] To assess the extent of this bias, we used the equation provided by Wacholder,[23] which is valid in the context of linear regression models only, but allows the difference between the surrogate measure and the true exposure value (i.e., the error E in measuring the true value) to be correlated with the 'true' value:

$$\textit{Bias factor} = \frac{\sigma^2 + \phi}{\sigma^2 + 2\phi + \omega^2} \tag{1}$$

where σ^2 is the variance of the true exposure, ϕ is the covariance of the true exposure and E, and ω^2 is the variance of E. For instance, if equation (1) yields the value 0.80, then the bias associated with the use of the surrogate measure is negative (i.e., the slope between the health outcome and the exposure is underestimated by 20%). We also calculated the 95% confidence interval of the bias estimate (1) using a bootstrap method with 1,000 replications.

The statistical analyses were conducted using R 3.0.1 (2013 The R Foundation of Statistical Computing) and STATA version 12.1.

RESULTS

Summary of Time-Activity Logs

The estimated percentages of time spent at home, work/school, and for commuting during a 7-day week are shown in Table 3. Subjects who used public transportation (tram, bus, train) as their main travel mode (29% of the sample) spent almost 50% and 66% more time traveling as individuals who primarily used motorized transportation (32%) or active transportation (walking: 19%, bicycling: 18%) for their daily commutes, respectively. The difference in duration occurs because individuals who use public transportation for the greatest length of their daily commutes combine public transportation more frequently with active transportation and spend more time transferring from one mode to another than the rest of the population (Supplementary Information, Supplementary Table S1). In fact, those who used mainly public transportation spent an average of 15 min per day waiting at public transportation stops and/or changing travel modes.

The average daily commute duration and distance reported by the study participants were 49 min and 14 km, respectively. On average, subjects living and working (or attending school) in Basel-City (240 subjects) had the shortest commuting distances

Table 3. Time spent in microenvironments during a 7-day week (%).

Microenvironment	*Main travel mode*	*n (subjects)*	*Mean ± SD*	*Median*	*Min*	*Max*
Home[a]		680	75.8 ± 4.5	74.0	64.9	87.3
Work or school		680	21.8 ± 4.0	25.0	12.5	25.0
Commute[b]	Total	680	2.4 ± 1.6	2.0	0.1	10.1
	Walking	129	1.6 ± 1.2	1.5	0.1	7.7
	Bicycle	121	1.6 ± 0.9	1.5	0.2	6.2
	Motorized transportation	220	2.1 ± 1.2	2.0	0.2	8.1
	Public transportation	198	3.9 ± 1.6	3.7	1.1	10.1
	Other	12	1.5 ± 1.0	1.0	0.3	3.7

[a]Includes weekends. [b]Time spent in traffic for daily travel between home and work/school on weekdays.

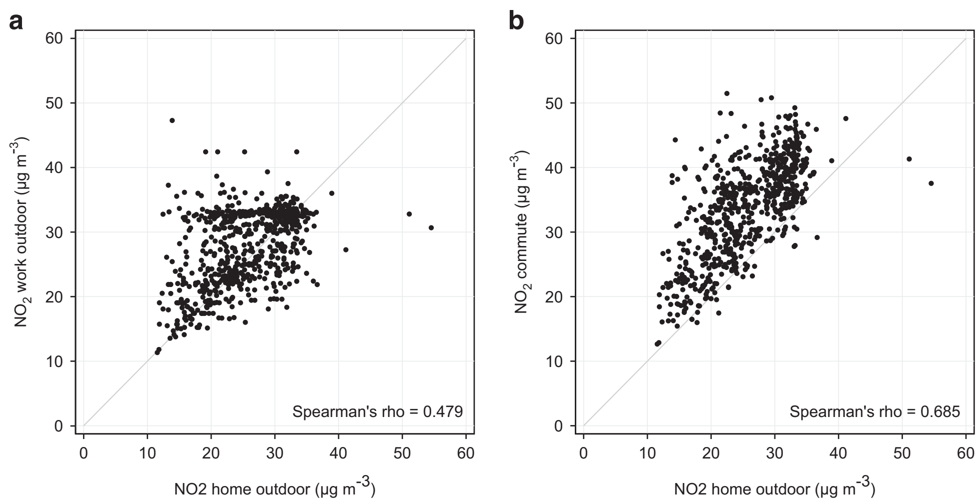

Figure 2. Comparisons between outdoor NO_2 concentrations at home and at work/school (**a**) and between outdoor NO_2 concentration at home and NO_2 concentration along the commuter routes (**b**).

compared with subjects living and working in Basel-Country (270 subjects) and subjects commuting between the two Cantons (170 subjects). The distances were 2.5- and 4-times longer for the latter subgroup than among subjects commuting within Basel-City and Basel-Country, respectively. The commute durations within Basel-City and Basel-Country were similar (42-43 min), but subjects commuting between the two Cantons spent 70 min on average in transit.

Residents commuting only within Basel-City primarily bicycled (30%), used public transportation (30%) or walked (27%). Motorized transportation was used by 9% of the participants. For commutes between Basel-City and Basel-Country, the subjects primarily used motorized travel (44%) and public transportation (46%).

Model Based Exposure Estimates

We observed higher mean (± SD) and more variable annual average NO_2 concentration estimates along commuter routes ($33.7 \pm 7.6\,\mu g\,m^{-3}$) than outdoors at work/school ($27.6 \pm 5.9\,\mu g\,m^{-3}$) and outdoors at home ($25.6 \pm 6.3\,\mu g\,m^{-3}$). Figure 2 shows scatterplots and Spearman correlation coefficients (ρ) between the outdoor concentrations at home and the concentrations at work/school and during commuting. In general, the correlations between these NO_2 concentrations were the strongest for subjects commuting within Basel-Country ($\rho > 0.5$). For subjects commuting between Basel-City and Basel-Country, we found the NO_2 concentrations at home and work/school to be negatively correlated ($\rho = -0.5$) (see also Supplementary Information, Supplementary Tables S2–S4). On average, the concentration during subjects' commute was 35% higher than that at the home address. The summary statistics for the time-weighted exposure and dose estimates used for the bias calculation (scenarios) are provided in Table 4. An additional table of subgroups (within Basel-City commuters, within Basel-Country commuters and between-Canton commuters) is provided in the Supplementary Information, Supplementary Table S5).

Contribution of Commute

Daily commutes between home and work/school contributed 3.2 ± 2.3% (range: 0.1–13.5%) to the overall weekly exposure. In comparison, the average contributions of home and work/school environments to total weekly exposure were 73.4 ± 7.4% and 23.4 ± 6.4%, respectively. Slightly higher contribution of the commute was observed when we adjusted for moderate (4.3 ± 3.0%, range: 0.2–17.3%) or high ventilation rates (5.4 ± 4.2%, range: 0.2–33.0%). Moreover, the average contribution of commuter exposure was almost twice as high for subjects commuting between Basel-City and Basel-Country (4.7 ± 2.4) compared with those commuting within Basel-City (2.6 ± 1.6) and within Basel-Country (2.8 ± 2.3) (Supplementary Information, Supplementary Table S6).

The contribution of the commute to total NO_2 exposure with and without adjustment for ventilation rates according to subjects' main commuter mode is shown in Figure 3 separately for the total study population and for subjects living and working (or attending

Table 4. Population NO_2 exposure (time-weighted over a 7-day week, $\mu g\, m^{-3}$) and weekly dose ($\mu g\, m^{-3} \times$ minutes $\times$ ventilation ratio) for the total study sample (n = 680 subjects).

NO_2 estimate	Mean ± SD	Min	p5	p25	Median	p75	p95	Max	iqr	Ratio p95/p5
H_{exp}	25.6 ± 6.3	11.5	15.0	20.8	25.4	31.1	34.0	54.5	10.3	2.27
H_{dose}[a]	258.0 ± 63.6	116.4	151.6	209.3	255.9	313.4	342.7	549.8	104.1	2.26
HW_{exp}[b]	26.1 ± 5.7	11.5	16.3	22.3	26.2	31.0	33.4	49.2	8.6	2.05
HW_{dose}[a,,b]	263.1 ± 57.0	116.1	163.8	225.2	264.0	312.0	336.7	495.6	86.8	2.06
HWC_{exp}	26.3 ± 5.6	11.5	16.5	22.5	26.5	31.2	33.5	48.8	8.7	2.03
$HWC_{dose\ moderate}$[a]	268.5 ± 57.8	116.2	166.7	229.4	269.6	319.4	343.7	499.7	90.9	2.06
$HWC_{dose\ high}$[a]	272.2 ± 60.6	116.2	167.2	231.2	273.9	324.9	355.4	529.1	93.7	2.12

Abbreviations: iqr, interquartile range; H, outdoors at home; HW, home and work; HWC, home, work/school and commuting; p, percentile. [a]Dose estimates are shown in 1000 $\mu g\, m^{-3}$. [b]Average commute exposure is assumed to be equal to the average outdoor exposure at home.

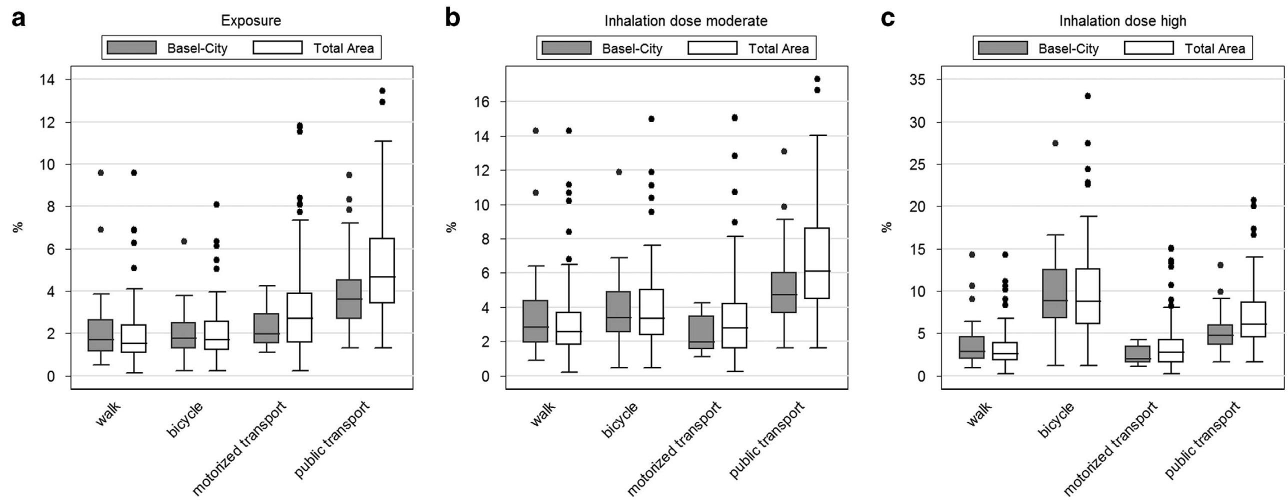

Figure 3. Contribution of commute (in %) to total weekly NO_2 exposure (**a**) and moderate (**b**) and high NO_2 dose estimates (**c**) for the entire sample (total area) and for subjects living and working in Basel-City by main commuter travel mode. Boxes represent the 25th to 75th percentiles, central lines represent the medians, bars stretch to the most extreme values within a distance from the box of less than 1.5 times the box length (i.e., the interquartile range), and dots represent the values that exceed the upper 75th percentile by more than 1.5 times the box length.

school) within Basel-City. For the whole-study area, median commute contribution to total exposure was the highest for subjects using mainly public (4.7%) and motorized transportation (2.7%) and the lowest for bicycle users (1.7%) and pedestrians (1.5%). For the ventilation-adjusted NO_2 concentrations, median commute contribution increased among public transportation users (6.1%), bicycle commuters (3.4%) and pedestrians (2.6%) and was the highest for bicycle users (8.8%) assuming high ventilation rates. The contribution of commute among subjects who walk to work/school within Basel-City was on average 0.2% higher compared with the total sample. Among commuters using motorized or public transportation, however, the percentage that the commute contributed to total NO_2 exposure and dose estimates was lower than that in the total population.

Scenario-Based Bias Results

The bias factor resulting from using only home outdoor NO_2 exposure estimates (H_{exp}) compared with using separate exposure estimates for home and work/school (HW_{exp}) was 0.88, indicating 12% attenuation bias (scenario 1) (see Table 5). We found similar bias results when comparing exposure and dose-adjusted models based on NO_2 estimates taken from outdoor locations at home only (H_{exp}, H_{dose}) with models using NO_2 estimates from home, work/school and the daily commute (HWC_{exp}, $HWC_{dose\ moderate}$) (scenarios 2 and 3). The attenuation bias weakened slightly (0.91) when we assumed a high ventilation rate for the travel legs completed by bicycle in the dose-adjusted scenario 4 ($HWC_{dose\ high}$). No significant bias was observed for scenarios 5 and 6, which ignored exposure or assumed a moderate commuter dose but did not ignore exposure at work/school. However, the health-effect estimates would be significantly overestimated by 4% using the NO_2 dose at home and work/school (HW_{dose}) versus estimates that also incorporate a commuter dose at a high ventilation rate ($HWC_{dose\ high}$) (scenario 7).

The bias results for the subgroups (within-Canton and between-Canton commuters) are provided in the Supplementary Information, Supplementary Table S7. We observed stronger attenuation biases associated with using home-only estimates (scenarios 1–4) for between-Canton commuters (< 0.70) than for individuals living and working within Basel-Country (between 0.87 and 0.89) and within Basel-City (between 0.82 and 0.83). In the total population, we found no significant underestimation of an effect estimate when we omitted the time spent in transport (scenario 5), whereas ignoring commute exposure produced a

Table 5. Estimated bias factors by scenario for the total study sample ($n = 680$ subjects) and Spearman's correlation coefficients between the two NO_2 estimates (ρ).

Scenario	*Simplified estimate*	*Assumed correct estimate*	*Bias (95% CI)*	*P-value*
1	H_{exp}	HW_{exp}	0.88 (0.86,0.89)	0.977
2	H_{exp}	HWC_{exp}	0.87 (0.85, 0.89)	0.972
3	H_{dose}	$HWC_{dose\ moderate}$	0.89 (0.86, 0.90)	0.969
4	H_{dose}	$HWC_{dose\ high}$	0.92 (0.89, 0.93)	0.952
5	HW_{exp}	HWC_{exp}	0.99 (0.99, 1.00)	0.999
6	HW_{dose}	$HWC_{dose\ moderate}$	1.01 (1.00, 1.02)	0.996
7	HW_{dose}	$HWC_{dose\ high}$	1.04 (1.03, 1.06)	0.979

Abbreviations: H, outdoors at home; HW, home and work; HWC, home, work/school and commuting.

significant 4% bias toward the null in the subgroup of individuals commuting between Basel-City and Basel-Country.

In general, a bias toward the null was observed when the surrogate measure had a larger range and showed more variability (expressed in the interquartile range or in the ratio between the 95th and 5th percentile) than the more refined measure to which it was compared (Table 4 and Supplementary Table S5). Computing time-weighted averages between the concentrations of the spatially well-dispersed home locations and those of the less-dispersed work and school locations (see map Figure 1) and travel routes decreased the variability. Instead, a positive bias emerged if the surrogate measure had a smaller range than the more refined measure to which it was compared. In addition, we found larger and more significant bias for scenarios and subgroups with weaker correlations between the NO_2 concentrations at home, work/school and during commuting.

DISCUSSION

We used time-activity data, including detailed information on travel routes of a representative sample in the area of Basel, to study the contribution of daily travel between home and work/school to total NO_2 exposure. The average time spent in traffic was 49 min per day, equivalent to 3.2% of the total exposure during weekly activities. Work or school occupied 22% of subjects' time on average. Ignoring time spent at work/school and related NO_2 exposure (i.e., using only outdoor exposure at home) would have resulted in a significant 12% underestimation of health effects. This bias was substantially stronger for subjects commuting between Basel-City and Basel-Country (33% underestimation) than for subjects commuting within those areas, underscoring the advantage of integrating at least home and work/school outdoor concentrations in long-term exposure assessment. In contrast, including commuter exposure in addition to home and work/school exposures had a negligible effect on NO_2 exposure estimates in the total population (Table 5). The relevance of commute exposure, however, was more relevant among subjects with longer commute distances—that is, those traveling between Basel-City and Basel-Country—but the related potential bias remained still very small (4%).

Although the relative contributions of commuter exposure and work/school exposure to total exposure were rather small, our data confirm that the impact on health-effect analyses may be relevant when both exposures are ignored, as done in most epidemiological studies, which are usually based on home outdoor measures alone. Our findings confirm results from other scenario-based modeling studies that human activity patterns may have an important role in air pollution exposure estimates. Dhondt et al.[4] used an activity-based transportation model to estimate the impact of NO_2 air pollution exposure on years of life lost due to respiratory mortality in Flanders and Brussels, Belgium. The predicted mortality rate increased by 1.2% when NO_2 estimates integrated both home outdoor and time-activity information. Their commuter exposure estimates were limited because it was calculated as the hourly average concentration on the whole road network, ignoring spatial differences. A study by Setton et al.[5] estimated the bias associated with omitting time-activity patterns using simulated NO_2 exposures from a microenvironment simulation of 382 census tracts (including an in-transport microenvironment assuming car travel) in Vancouver and NO_2 exposures of spatially and temporally linked activity patterns (including routes between origin-destination points modeled as straight lines) in Southern California.[14] Using the same bias estimation method as our study, they reported similar bias (0.84 for the metropolitan area of Vancouver and 0.93 for Southern California). A stronger negative bias (0.70) was found for Vancouver when the spatial variability of the air pollution model increased.

Our analyses reveal that time spent at work or school has a stronger impact on total NO_2 exposure than daily commute. This holds true even in models adjusting for increased ventilation rates during active commute. A higher relevance of work than commute on total exposure has also been reported in short-term NO_2 personal monitoring studies[6,8,16] and modeling studies that extract exposure information from air pollution models using GPS data. For example, Nethery et al.[7] reported improvements in predicting NO_2 personal exposure data for 38 pregnant women in Vancouver when using a combined home and work estimate from a NO_2 land use regression (LUR) model. Adding transit-based LUR estimates extracted from GPS data had little additional effect on the exposure estimates. However, in Barcelona, time spent in transit (6% of total time) contributed on average 11% and 24% to the total daily modeled NO_2 exposure and inhalation dose, respectively, among 36 adult subjects.[3] The study by de Nazelle et al.[3] used physical activity data and geographic location data from smartphones. Higher in-transit NO_2 levels, longer commute times and greater contrasts between street environments and urban background likely contributed to the higher commute contributions in Barcelona than in our study. Similarly, we found that the strongest bias toward the null was associated with neglecting work/school locations for subjects commuting between the urban center of Basel-City and the rural to suburban surrounding area of Basel-Country. The exposure misclassification is likely explained by the greater differences in NO_2 concentrations between home and work/school, as illustrated by the negative correlation, and as Setton et al.[15] have also demonstrated. For the same population subgroup, ignoring the time spent in traffic also contributed significantly to the underestimation of NO_2 exposure. This finding indicates that for people with longer in-transit time inclusion of exposure during commute may be advisable. In addition, the high proportion of motorized transport (44%) commuters likely contributed to this finding. Air pollution exposure along car routes, which mainly follow major roads, is higher than the exposure associated with other routes and trips using other travel modes in the study area.[9,20]

We observed considerable differences in bias factors when we used dose-adjusted estimates based on high ventilation rates compared with estimates based on moderate ventilation rates. Assuming increased ventilation rates during active commuting led to higher commute contributions for dose-adjusted NO_2 exposures. Under the extreme assumption of applying high ventilation rates to the entire bicycle commute, the proportion of total dose being commute-related ranged up to 33%. Interestingly, under the assumption that this high-dose commute model would reflect the best estimate of the unknown truth, our analysis indicates that a model based only on the combination of home and work/school concentrations may overestimate the effects (scenario 7). Our dose estimates are neither based on actual physical activity measures nor on physiological characteristics of the participants. As our approach reflects the average exposure misclassification and related bias in health-effect estimates for a random population sample, misclassification may be different on an individual level. Thus, while our data may be used for population-based risk assessments, an individual risk assessment could only be derived if more accurate information on inhalation parameters while commuting and at other places was available. While this was not the purpose of our study it needs to be considered if one would like to translate the findings to individuals.

To our knowledge, this is the first study estimating the contribution of commuter exposure to total NO_2 exposure according to the main mode of travel for a large, representative population sample using actual travel route data for each leg of a trip and information about the waiting time between legs. We found that the commute produced the highest contribution to total NO_2 exposure for subjects who mainly used public transportation. This result is likely due to the combination of active transportation modes and the considerable amount of time spent at public transportation stops. In dose-adjusted NO_2 models, commute contributions were the highest for bicycle and public transportation commuters. The comparison between travel modes based on legs only revealed the highest NO_2 exposures along the legs of the trips using motorized transportation.[20] Most previous comparisons of air pollution exposure according to travel mode—for example, de Nazelle et al.,[3] Ragettli et al.[9] and Int Panis et al.[13]—were performed along pre-defined trips and did not take into account possible combinations of travel modes. In addition, changing transport at locations with potentially high exposure levels (e.g., at bus stops along busy streets) is often not considered in those studies. These factors seem to be important to consider in policy initiatives promoting the use of public transportation and reducing reliance on private motor vehicles.

Our findings may be limited to populations living and working within a small study area like Basel. Stronger bias toward the null may be expected when subjects with longer commute distances are included.[4,5] This attenuation bias may be especially strong for subjects traveling between areas and locations characterized by considerable contrast in air pollution concentrations. Our ability in estimating these contrasts were likely limited by the 100 m spatial resolution of the air pollution model used.[5] Nevertheless, we were able to show spatial differences in exposure levels across different time-activity patterns by comparing population subgroups commuting within and between Basel-City and Basel-Country. Moreover, our NO_2 exposure estimates are based on home and work/school locations only as people usually spent most of their time at those places. Similarly, we only included travel related to work and school activities because those commutes belong to the most important travel reasons and are carried out regularly over a certain time period. Additional bias introduced by not including other places and travels is likely to be rather small and most likely non-differential, thus resulting in some additional bias toward the null. However, on an individual level, other activities that are carried out on a regular basis and for a considerable amount of time may have an effect on total exposure, especially when taking place at areas with lower or higher air pollution concentrations than at the others. Repeated 1-day travel records would be needed to investigate the effect of other travels and activities. Further limitations of our analysis include that we did not consider in-cabin modification and differences in NO_2 concentrations between in-transport microenvironments as for example the study in Barcelona by de Nazelle et al.[3] due to the absence of such NO_2 data for the study area. However, while these factors add to the misclassification of commuter exposure estimates, our findings indicate that these factors would be of rather minor influence on total NO_2 exposures from outdoor sources.

Given our focus are epidemiological studies on the health effects of outdoor air pollution, our investigation ignores indoor sources of air pollution. We use NO_2 strictly as a marker of ambient —in our case mostly traffic-related—air pollution. As in most epidemiological studies on long-term health effects, outdoor concentrations are used as proxy for exposure,[1] although true exposure to NO_2 from outdoor sources could be calibrated during the time indoors using indoor/outdoor ratios. This is not the purpose of this analysis as our findings should remain relevant for the interpretation of epidemiological studies on outdoor concentrations. Furthermore, NO_2 serves only as an indicator of the complex mixture of harmful traffic-related air pollutants whose composition may vary spatially and temporally. As a proxy for the traffic pollutant mixture, one would expect to find varying associations of NO_2 with other health-relevant pollutants across the city.[10] Nevertheless, NO_2 is undoubtedly a traffic-related pollutant and we have shown that bias occurs when we only consider the home outdoor exposures. To assess the degree of bias for other pollutants, similar simulations are needed for other traffic-related air pollutants, such as ultrafine particles and black carbon. For these primary vehicle exhaust emissions, the bias that occurs when commuter exposures are ignored is possibly stronger because they have greater spatial heterogeneity than NO_2.[7]

Although these findings may not be generalizable to all studies on the long-term effects of traffic-related outdoor air pollution on health, they are certainly relevant for the Swiss SAPALDIA study. Our assessment indicates that the previously published results—all based on home outdoor exposure only—to inherently though not excessively underestimate associations. While the simple home outdoor models were sufficient in the past, the ever-decreasing levels of air pollution and the shrinkage of spatial contrasts in concentrations seen in Switzerland, and other countries where clean air policies have improved air quality, may call for the adoption of exposure models that integrate at least outdoor concentrations at home and work/school if not during commute. Otherwise, the impact of non-systematic exposure misclassification may jeopardize the ability to detect long-term effects of ambient air pollution. We showed that the potential effect of including outdoor exposures at work/school locations and related transit patterns in NO_2 exposure assessment to investigate long-term health effects depends on commute distances between home and work/school locations, prevalent commute modes and the spatial contrast of air pollution concentrations.

CONFLICT OF INTEREST

The authors declare no conflict of interest.

ACKNOWLEDGEMENTS

This work is part of a Europe-wide project, Transportation Air Pollution and Physical Activities (TAPAS), which is an integrated health risk assessment program on climate change and urban policies. The study is partially supported by the Swiss National Science Foundation (SNSF 324730_135673). We acknowledge the Federal Office for the Environment for supplying the fixed site measurement data and the NO_2 model

PolluMap. We are thankful to the Federal Statistical Office of Switzerland for providing the census data.

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Supplementary Information accompanies the paper on the Journal of Exposure Science and Environmental Epidemiology website (http://www.nature.com/jes)

5 SUMMARY OF MAIN FINDINGS

In general, smaller average particle sizes and higher UFP levels were measured at places and for transportation modes in close proximity to traffic (article 1, chapter 3). Sidewalk UFP concentrations were higher in the densely populated urban residential area and in the town centre of Basel metropolitan area than in the green residential area with less traffic density. Along the same main road, UFP concentrations were highest in car and on the bicycle. During walking and in tram, UFP concentrations were on average 40% lower than in car. UFP concentrations were lowest in bus, with almost 60% lower concentrations than inside the car. Concentrations were highest for all transportation modes during weekday morning rush hours compared to non-rush hours and weekends. Average particle size in the five transportation modes ranged between 46 and 51 nm. Bicycle travel along main streets between home and work place (24 min on average) contributed 21% and 5% to total daily UFP exposure in winter and summer, respectively. Contribution of bicycle commutes to total daily UFP exposure could be reduced by half if main roads were avoided. The average contribution of commute to total daily exposure along the high exposure route over all six measurement weeks (covering summer, winter and spring) was 8%.

Within Basel-City, estimated average time-weighted NO_2 population exposure during commuting was similar among all air pollution models (around 39-41 µg m^{-3}) (article 3, chapter 4). Within-city and within-subject variability in annual mean NO_2 commuter exposure with the high resolution dispersion model (grid size 25 m) was larger than with the dispersion model with a lower resolution (grid size 100 m) and the LUR model (applied to a 50x50 m grid). Enlarging the study area to the surrounding area of the city, commuter NO_2 estimates from the dispersion model with the lowest resolution showed greater variability than just within Basel-City. Median NO_2 exposures were highest along motorized transportation and bicycle legs (i.e., pieces of the trips with the same transportation mode) and lowest for walking.

The population working (>= 50% work load) or attending school within the region of Basel spent on average 49 minutes for daily commutes, which was equivalent to 2.7% (range 0.1-13.5%) of the total exposure during weekly activities (article 3, chapter 4). Slightly higher median contribution rates were observed when we considered moderate (3.5%, range: 0.2-16.8%) and high ventilation rates (4.2%, range: 0.2-33.4%) during active transportation. On average, the NO_2 concentration during subjects' commute was 35% higher than at the home address. Work or school occupied 22% of the subjects' time on average. Ignoring the time spent at work or school and the related NO_2 exposure (i.e., using only outdoor exposure at home) would have resulted in a significant 12% underestimation of health effects. This bias was even stronger for the subjects commuting between Basel-City and the rural to suburban surrounding area of Basel-Country (33% underestimation) than for the subjects commuting within those areas. For the same population sub-group, we observed a potentially significant underestimation of health effects (5%) attributable to including outdoor exposures at home and at work/school but omitting exposure during the commute.

6 GENERAL DISCUSSION

We showed that measured UFP concentrations and simulated NO_2 concentrations are generally higher in traffic microenvironments than at urban background locations and at home and work places in the absence of indoor sources. Our results are in agreement with other studies showing that the transportation mode is a significant determinant of traffic-related air pollution concentrations to which people are exposed during daily travels (de Nazelle et al., 2012; Knibbs et al., 2011). However, comparing the results of the UFP monitoring study in Basel to other studies illustrates that UFP concentrations and the transportation mode with the highest and lowest UFP concentrations differ (Table 6-1). There are several factors that make a uniform ranking of transportation modes in order of UFP concentration levels difficult. These factors will be discussed below based on the components *source* and *concentration* of the exposure concept described in chapter 1 (Figure 1-1).

6.1 Determinant factors of UFP concentration levels in transport microenvironments

Sources of traffic-related air pollution

First of all, the strength and characteristics of the source, i.e. of traffic-related air pollution, varies across study settings. Traffic factors such as traffic intensity and composition of the traffic fleet (gasoline-powered, diesel-powered and heavy-duty vehicles) differ between cities and countries. UFP emissions of diesel engines can be about two orders of magnitude greater than those of gasoline engines (Beddows and Harrison, 2008; Kittelson et al., 2004). These reasons partially explain the higher UFP concentration levels measured in various transportation modes, for example in London (Kaur et al., 2005) and Barcelona (de Nazelle et al., 2012). As shown in the Basel commuter UFP study, traffic density is also a significant determinant in UFP concentration levels within cities and between roads.

Table 6-1. UFP personal monitoring studies with multiple transportation modes.

Study	Location	Routes of transportation modes	Design – Sampling periods	Transportation modes	Mean UFP (particles cm^{-3})	Instrument used for UFP measurements	Other air pollutants measured
Current study Ragettli et al. (2013)	Basel, Switzerland	Same main route	3 periods: weekday rush hours, weekday non-rush hours, weekends March–May, September	Walking Cycling Bus (diesel & CNG) Tram Car (gasoline)	19,500 22,700 14,100 18,800 31,800	MiniDiSC	Average particle size
Kaur et al. (2005)	London, United Kingdom	Street canyon intersection, partly separate routes for walking and bicycle	3 periods: morning, lunch, afternoon	Walking Bicycle Bus (diesel) Car (gasoline) Taxi	67,800 94,000 101,400 99,700 87,500	P-Trak	$PM_{2.5}$, CO
Briggs et al. (2008)	London, United Kingdom	Nearly same routes	Simultaneously driven by car until walk was completed weekdays May–June	Walking Car (diesel)	30,300 21,600	P-Trak	PM_1 Fine PM ($PM_{2.5}$-PM_1) coarse PM (PM_{10}-$PM_{2.5}$)
Weichenthal et al. (2008)	Montreal, Canada	Different routes	2 periods: morning and evening rush hours Three seasons from April–November	Walking (includes waiting for bus) Bus Car (gasoline)	20,500 25,300 34,900	P-Trak	
Boogaard et al. (2009)	11 cities in Netherlands	12 routes in each city, separate routes for transportation modes	Simultaneous measurements August-October, 12pm–19pm	Bicycle Car	24,300 25,500	CPC3007	$PM_{2.5}$

Study	**Location**	**Routes of transportation modes**	**Design – Sampling periods**	**Transportation modes**	**Mean UFP** (particles cm^{-3})	**Instrument used for UFP measurements**	**Other air pollutants measured**
Int Panis et al. (2010)	Belgium: Brussels Louvain-la-Neuve Mol	Same route	Bicycle was measured after the car ride June, no information on sampling time	Bicycle Car Bicycle Car Bicycle Car	30,200 31,500 11,900 11,600 8,700 14,200	P-Trak	$PM_{2.5}$, PM_{10}
Knibbs and de Dear (2010)	Sydney, Australia	Nearly same routes for car, bus, train	Morning and evening rush hours September–October	Bus (diesel & CNG) Car (gasoline) Ferry Train	105,000 89,000 55,000 46,000	CPC3007	$PM_{2.5}$
Zuurbier et al. (2010)	Arnhem, Netherlands	Same routes, plus a low exposure bicycle route	8–10am weekdays, repeated measurements over one year Simultaneous measurements in pairs (within same mode)	Bicycle, high-traffic Bicycle, low-traffic Diesel bus Electric bus Car, diesel Car, gasoline	48,900 39,600 43,200 28,600 37,100 40,500	CPC3007	$PM_{2.5}$, PM_{10}, soot
de Nazelle et al. (2012)	Barcelona, Spain	Nearly same route	5 periods: morning and afternoon rush hour, lunch, morning and evening non-rush hour, pairwise measurements, May–June	Walking Bicycle Bus Car (diesel)	52,700 77,500 55,200 123,000	CPC3007	CO, CO2, BC, $PM_{2.5}$
Quiros et al. (2013)	Santa Monica, California, USA	Same route	3 periods: morning rush hour, evening rush hour, morning non-rush hour	Walking Bicycle Car, windows open Car, windows closed	17,600 18,600 18,600 4,800	CPC3007	$PM_{2.5}$
Both et al. (2013)	Jakarta, Indonesia	Different routes	May–October	Bus Car	401,000 294,000	CPC3007	$PM_{2.5}$, CO

Proximity to the air pollution source

Based on the spatial characteristics of traffic-related air pollutants, it would be generally expected that transportation modes closest to freshly emitted vehicle exhaust (i.e. the source) are characterized by the highest air pollution levels. Hence, people in on-road vehicles would be exposed to higher concentrations than cyclists on the side of the road, followed by pedestrians on the side walk. However, this was not the case in the Basel UFP monitoring study. Over the entire monitoring period, the median UFP concentrations adjusted by a fixed site station were similar both in car and on bicycle. Looking at different times of the day and week, UFP levels during weekday morning rush hours were higher on bicycle than in car. The opposite was true for weekday afternoon rush hours and non-rush hours.

In previous personal monitoring studies, the proximity to the source did not uniformly determine the mode in which the highest concentrations were recorded (Table 6-1). A study in three Belgian cities *compared the same urban routes* for motorists and cyclists and found no significant differences in UFP concentrations during bicycle trips immediately following car trips in two of three cities. In the third city, which was the smallest city among the three, UFP concentrations were about 60% higher in cars than while riding bicycles (Int Panis et al., 2010). Up to 70% lower UFP concentrations during car driving versus cycling and walking were observed in a study in Santa Monica, California, which measured UFP concentrations simultaneously along the same route. In the same study, no relevant differences in UFP concentrations were reported when windows were open rather than closed with air conditioning recirculation applied (Quiros et al., 2013). Another similar study with a pair-wise design in Barcelona reported around 50% and 40% higher mean UFP concentrations in cars with open windows compared to walking and cycling, respectively (de Nazelle et al., 2012).

In studies that measured UFP concentrations *along different roads*, i.e. each transportation mode followed another route, UFP concentrations were mostly reported higher in car or bus than during walking and cycling, especially when active transportation modes followed a road with less traffic (Boogaard et al., 2009; Kaur et al., 2005; Weichenthal et al., 2008).

More consistent reporting exists for characterization of the peaks in UFP concentrations. High and short peaks, lasting only for a few seconds, have been generally observed during cycling, whereas in cars fewer and lower peaks of longer duration occur (Boogaard et al., 2009; Int Panis et al., 2010; Kaur et al., 2006; Zuurbier et al., 2010). As also observed in the Basel study, high peaks during cycling are mainly attributed to passing buses, trucks and motorcycles or to congested traffic at stop lights. High-emission preceding vehicles and the mixing of air pollutants with in-cabin air likely explain the longer peaks in cars. Averaging the measurements by minute or longer time periods generally results in higher peaks in cars than during cycling (Boogaard et al., 2009; Zuurbier et al., 2010). We computed median UFP concentrations per transportation mode and trip to minimize the influence of one, or a few, unusually heavy emitters passing by while monitoring and to ensure representativeness for the general commuting situation (the difference between mean and median concentrations is also discussed in article 1). This approach could be questionable if very short high peak exposures were of particular health relevance as compared to distributing the same dose of pollutants over a longer time period. However, to date there is no evidence for a particular health relevance of short peaks, which supports keeping the focus on integrated estimates.

Meteorology

On- and near-road UFP concentrations are further determined by location-specific meteorological variables. Temperature, wind speed and wind direction are the most frequently reported. In-transit studies have reported negative correlations between temperature and UFP concentrations (correlation coefficients around -0.76), with stronger relationships for cycling than for automobiles (Knibbs et al., 2011). This likely also explains the higher median UFP concentrations during cycling than during car driving in the cooler morning rush-hours in Basel. Higher wind speed is usually associated with more dilution of particles resulting in lower concentrations (Kaur and Nieuwenhuijsen, 2009; Knibbs and de Dear, 2010). Comparisons of transportation modes and respective concentrations are constrained as measurements have been conducted during various times of the day, week, and year. It is well known from fixed-site measurements that UFP show diurnal, weekly and seasonal patterns typical of temporal patterns of traffic density and meteorological conditions. In urban areas, highest ambient UFP levels are usually observed during morning weekday rush hour with a second, less distinct, peak during afternoon rush hours (Borsós et al., 2012; Morawska et al., 2008). This was also seen in Basel (Figure 4 in article 1). Not all studies carried out measurements within the same time period in the different transportation modes (e.g. Boogaard et al., 2009; Int Panis et al., 2010) and therefore relative differences in UFP concentrations were potentially misclassified. To the best of my knowledge, our study was the first multimodal in-transit study to include a weekend sampling period. We found less contrast in UFP concentrations between modes during weekend than during weekday rush and non-rush hour.

Built environment

Furthermore, various factors have been suggested to affect the proportion of UFP concentration that directly comes into contact with people in transport environments. These include characteristics of the built environment such as building infrastructure and road layout (Boarnet et al., 2011; Buonanno et al., 2011). We did not study the effect of positional factors, for example, separations from vehicles by parked cars, trees or separate bicycle lanes, on cyclists' and pedestrians' UFP exposures. Previous studies have shown mitigating effects of such street designs (Kendrick et al., 2011). The route in Basel included marked on-road cycling lanes and sections where pedestrians were separated from the street by parked cars or trees. The most substantial within-mode variability, however, has been reported for cars and buses. In fact, measuring and understanding in-cabin UFP concentrations within motor vehicles is a complex and unique field of research.

Cabin ventilation

A key determinant of in-cabin UFP concentration is ventilation (Bigazzi and Figliozzi, 2012; Fruin et al., 2011; Hudda et al., 2012; Knibbs et al., 2011). As mentioned above, open windows can significantly increase UFP levels in cars, even reaching in-cabin/on-road (I/O) UFP ratios of 1 due to higher air exchange rates (Hudda et al., 2011). Maximum in-cabin protection (I/O ratios between 0.02-0.39) was observed when windows were closed and ventilation set to re-circulate in-cabin air (Hudda et al., 2011; Knibbs et al., 2010; Zhu et al., 2007). However, this ventilation setting is rarely applied as it can lead to very high accumulation of CO_2 (Tartakovsky et al., 2013). Most personal monitoring studies including car driving in Northern and Northwestern Europe (also in Basel) applied ventilation settings with closed windows, air conditioning turned off and ventilation fan system (outside-air intake) set to moderate level. A recent study in Guildford, a midsized town in the United Kingdom (UK), reported a mean I/O ratio of 0.72 under the same ventilation characteristics. I/O ratios (0.55) were smaller for particles in the size range 5-30 nm than for particles 30-300 nm (0.82). The reduction of the smaller particles in cars relative to outdoor air is likely due to coagulation of smaller particles in the ventilation system (Joodatnia et al., 2013). Knibbs et al. (2010) showed that reducing the fan velocity potentially further reduced particle filtration efficiency for all size ranges. Hudda et al. (2011) concluded, after comparing several cars with different ventilation characteristics, that on-road and in-cabin size distributions are very similar and on-road size distribution does not necessarily affect I/O ratios of total particle number counts. Additional factors that have been reported to affect in-cabin particle concentrations include age of the car and driving speed. In older cars, reduced sealing efficiency of windows and doors, facilitating in-cabin particle penetration, was observed when compared to newer cars. With faster driving speed, increased air exchange rates between roadway and in-cabin air was found (Fruin et al., 2011; Hudda et al., 2011).

We found smaller mean and median particle number concentration and larger average particle sizes for bus compared to car travel. UFP concentration was lowest in bus, regardless of time period. This is likely explained by the different ventilation settings and less infiltration of smaller particles in the well-encapsulated bus cabins. Self-pollution can be a significant source of air pollution in the cabin, even when windows are closed (Behrentz et al., 2004; Liu et al., 2010). There is evidence of fuel type and emission control devices influencing in-bus UFP concentration levels. In a comparison by Knibbs et al. (2011) of trip-weighted mean UFP concentrations measured in buses in various studies, lowest mean (±standard deviation) UFP concentrations were recorded for buses powered by compressed natural gas (CNG) (17,000 ±8000 particles cm^{-3}), and highest were recorded in diesel buses (48,200 ±3200 particles cm^{-3}). A reduction of about 50% was observed in diesel buses equipped with particulate filters relative to diesel buses without an emission control device (Knibbs et al., 2011). In Basel, the bus fleet was composed of 60% diesel-powered vehicles, all of them equipped with particulate filters. The other 40% of the buses were powered by CNG. The buses were relatively new, with most of them put into operation after the year

2000 (personal communication with *Basel Verkehrsbetriebe BVB*, 2013). The number of measurements was too small to study an effect of fuel type. However, average UFP concentrations measured in buses in Basel (14,000 ±8000 particles cm^{-3}) are consistent with the levels reported by Knibbs et al. (2011).

Concentration differences due to sampling equipment

Finally, comparisons between different studies might be impaired as in-traffic exposures are measured with different monitoring equipment (see Table 6-1). Commonly used particle counters in personal in-transit studies are the P-Trak, which starts counting particles from 20 nm, and the condensation particle counter CPC3007, detecting particles of 10 nm and larger. Vehicle exhaust can have large fractions of particles with diameters smaller 20 nm (Westerdahl et al., 2005). Therefore, studies that have used P-Traks potentially underestimated personal commute UFP levels (Zhu et al., 2006). The miniature Diffusion Size Classifier (miniDiSC) which was used in this study (now commercially available under the name DiSCmini) measures particle concentration and average particle size distribution diameter in the size range of 10-300 nm. In collocated comparisons, a lower accuracy was found for the miniDiSC (±30%) than for CPC 3007 (±5%) (Asbach et al., 2012). However, both CPC3007 and the P-Trak have some practical short-comings compared to the customized miniDiSCs used for this thesis work, mainly due to their bigger size, shorter battery life, dependence on working fluids and the requirement to maintain them in a horizontal position. Therefore, in contrast to the miniDiSC, P-Trak and CPC3007 are not as useful for personal monitoring studies with multiple individuals, as it is common in epidemiological studies.

In summary, transportation mode is a significant determinant of the UFP concentration to which people are exposed during daily travels. There are various factors affecting UFP concentrations within and between transportation modes. Attempts to quantify those determinants are scarce (Kaur and Nieuwenhuijsen, 2009). It may not be adequate to generalize findings across different traffic conditions and ventilation settings. Factors likely differ by geographic location. Therefore, personal in-transit studies in diverse areas are needed to accurately understand commuter exposure to UFP. Unfortunately, to date, only two UFP commuter studies outside Europe and Northern America have been conducted, one in autorickshaws in New Delhi, India (Apte et al., 2011) and one in commuters in Jakarta, Indonesia (Both et al., 2013).

6.2 Correlation of traffic-related air pollutants in transport environments

In the personal monitoring study in Basel, only UFP as a marker of traffic-related air pollution was described. One could raise the question of whether UFP in-transit measurements can be used to predict other traffic-related air pollution or vice versa. A number of personal in-transit studies measured several pollutants at the same time (see Table 6-1). In general, the correlation of UFP with $PM_{2.5}$ and PM_{10} are low and non-significant. There is also no clear relationship between the strength of the relationship and the transportation mode. Some studies have observed smaller modal contrast for $PM_{2.5}$ than UFP (de Nazelle et al., 2012; Knibbs and de Dear, 2010; Zuurbier et al., 2010). This likely reflects differences in the nature of the sources of smaller and larger particles (Morawska et al., 2008) and the lack of a simple linear relationship between particle number and particle mass emissions from vehicles (Zhu et al., 2008). In addition, comparisons between fixed site measurements along roads and the urban background showed larger gradients for UFP than $PM_{2.5}$ and PM_{10}, illustrating a more homogenous distribution of PM in urban environments (Boogaard et al., 2010; Zuurbier et al., 2010).

A somehow stronger relationship with UFP would be expected for BC since vehicle exhaust is the common and major source in urban streets. Zhu et al. (2002) showed that those pollutants undergo similar dispersion processes and track each other well with increasing distance from a major highway. Westerdahl et al. (2005) reported a Spearman correlation coefficient of 0.88 between UFP and BC in on-road measurements using a mobile platform in Los Angeles. The relationship between UFP and BC is generally stronger for diesel than gasoline engine emissions (Zhu et al., 2008). Nevertheless, short-term personal monitoring studies in multiple transportation modes reported weak correlations (0.1-0.5), especially when conducted inside vehicles (de Nazelle et al., 2012; Zhang and Zhu, 2010; Zhu et al., 2008).

There is also no evidence for a consistent relationship between in-transit UFP and NO_2 (Knibbs et al., 2011). NO_2 is not only emitted directly by vehicular fuel combustion, but also secondarily formed in the atmosphere by NO and ozone (O_3). A somehow stronger correlation between UFP and NO than with NO_2 was observed on roadways (Beckerman et al., 2008). Concentrations of NO_x (including both NO_2 and NO) – and related correlation with UFP – in urban streets depend on various factors including appropriate meteorological conditions for the secondary formation of NO_2, availability of other sources of NO_2 (industry, shipping, heating), traffic intensity, the proportion of diesel-engine vehicles in the fleet and other vehicle-specific factors. In recent years, there has been a trend of increasing primary NO_2 in urban street environments while at the same time NO_x emissions have been decreasing. The reason for increasing primary NO_2 emissions is the more common use of diesel-powered cars which emit more NO_2 than gasoline-fuelled vehicles (Carslaw, 2005; Hueglin et al., 2006). Primary NO_2 constitutes less than 5% of total NO_x in the emissions of petrol-fuelled vehicles and 10-12% in diesel vehicles without modern exhaust treatment. The implementation of after exhaust treatment technology such as particle filters and oxidation catalysts further contribute to an increase of primary NO_2. Some catalyst-based particulate traps of diesel vehicles convert NO to NO_2 in the exhaust in order to promote the oxidation of collected soot in the filter (Grice et al., 2009).

Overall, correlations between UFP and other air pollutants during simultaneous in-transit measurements are inconsistent. Similar to the differences in UFP concentrations between means of transportation, transportation mode- and location specific parameters are suggested to be responsible for observed variability. This indicates that no surrogate can be used for UFP number concentrations for estimating commuter's exposure to UFP. In-transit studies generally show higher associations between traffic density and UFP concentration than for larger particles and particle mass (Briggs et al., 2008; Kaur and Nieuwenhuijsen, 2009).

6.3 The influence of travel time and route on commuter exposure

Following the exposure concept introduced in chapter 1, the actual time spent in traffic is necessary to estimate commuter's *exposure* to traffic-related air pollution. Within Basel-City, we observed the highest contribution of commute to total weekly NO_2 exposures for those people also spending the longest time in traffic (article 3, chapter 4). Highest contributions (median: 4%, range 1-9%) to total NO_2 exposure and longest travel durations (about one hour per day) were related to commuters using mainly public transportation. The lengthy in-transport durations could be explained by the frequent combination with active transportation and the considerable amount of time spent at public transportation stops (on average 15 minutes per day). The population sub-group with the largest contributions (median 5%, range 1-13%) of commute to total exposures, however, included people commuting between Basel-City and Basel-Country. Besides the effect of in-transit duration (on average 70 minutes per day) and distance, this finding is also explained by a high proportion of motorized transportation users (44%) following busy roads. The comparison between the transportation modes based on legs only revealed the highest NO_2 exposures along the legs of the trips using motorized transportation (article 2, chapter 4). For this sub-group, ignoring in-transit exposure would have resulted in significant underestimation of potential health effects (article 3, chapter 4). Hence, not only the duration, transportation mode and trip timing is important in estimating in-transport exposures to traffic-related air pollution but also the route. The determinant effect of the route is also shown in the personal monitoring study, where the contribution of commute to total UFP exposure could be reduced by half when choosing a bicycle route which avoided busy streets. The difference in exposure was observed even though the trip duration along the low-exposure route was about five minutes longer.

Previous studies have estimated the influence of in-transport UFP exposures on the population's total exposure without using 24-hour personal monitoring or detailed travel behavior data but rather using concentration data from the literature or stationary measurements. For example, a few have integrated in-automobile measurements to estimate average contributions for the population. Fruin et al. (2008) and Zhu et al. (2007) calculated an in-vehicle contribution to total daily UFP exposure of 33-45% and 10-50%, respectively, assuming that Americans spend on average 90 minutes in automobiles per day. Both in-car measurements were carried out on Los Angeles freeways and arterial roads, which are characterized by a high density of diesel-powered heavy-duty vehicles. Wallace and Ott (2011) estimated that driving the same amount of time along roads with fewer diesel trucks, between cities in California and on the East Coast of the USA, contributes on average 17% to total daily UFP exposure. In Jakarta, Indonesia, where on-road UFP concentrations are about two to three times higher than in the USA, the fraction of total time-exposure due to commuting by private car (three hours per day) was similar (25%) for UFP to those calculated for the USA (Both et al., 2013). The authors attributed this relatively low contribution, in view of the high in-transit UFP exposures, to proportionately higher UFP concentrations at home and work places.

6.4 Differences in short-term and long-term estimates of commute contributions to total exposure

The population-based modeled percentages of commute contribution to total exposure for NO_2 were smaller (median: 3%) than those of the 24-hour personal UFP measurements (median 8%). This difference is partly explained by the fact that those are two different pollutants which have been shown to not correlate well (see chapter 6.2). Additionally, the personal monitoring was carried out by only one person during six weeks over the year while population-based NO_2 exposure estimates are based on a map of annual mean NO_2. Potential further reasons for the lower percentages of time-exposure from commute for NO_2 than UFP and potential underestimation of the simulated NO_2 estimates are discussed in the following paragraphs.

Like any modeling analysis, our exposure simulation was based on some assumptions and had sources of uncertainty. Concentration simulations occurred on a dispersion model of a resolution of 100x100 m. NO_2 concentration of a leg of a trip was computed based on the distance-weighted averages of the underlying model grids. The model itself has sources of uncertainty related to methodology and the quality of input data. In article 2 (chapter 4), we evaluated the difference between modeled commuter concentrations when applying models with higher spatial scales. Average differences of time-weighed commuter concentrations within Basel-City between models were rather small (between 0.97-2.04 µg m^{-3}) but significant. However, even the scale of 25 m from the evaluated model with the highest resolution may be too coarse to assess the small-scale variability of on- and near-road exposures.

In the NO_2 exposure simulation, we did not take into account travel microenvironments such as in-vehicle exposure modification due to the potential use of ventilation systems or the commuter's position on the road. Therefore, we may have over- or underestimated in-vehicle NO_2 concentrations. This issue has given rise to several discussions. We explored various options including a) applying ratios between transportation modes of our UFP data, b) approximating the relationship between UFP and NO (which would require extensive data on the highly variable relationship between NO and NO_2 across the study area), c) approximating differences between transportation modes by the decrease of NO_2 or NO concentration with increasing distance from the road, or d) using data from the literature. However, as discussed in chapter 6.2, correlations between UFP and NO as well as NO_2 are highly variable and depend on several factors. We did not have the equipment for doing in-transit NO_2 or NO measurements. In our literature review on in-cabin NO_2 exposure, we found that information on relationships between in-cabin NO_2 and on-road NO_2 is very rare. To my knowledge, there is only one study, by Chan and Chung (2003), which measured the I/O relationship of NO_x while driving in Hong Kong. I/O ratios varied between ventilation systems and driving environment. Using the fresh-air ventilation mode, the I/O ratio of NO_2 changed from approximately 0.9 to 4.4 as the car commutes from a highway to the country-side.

Uncertainties of estimated NO_2 commuter exposure related to the quality and accuracy of geographic information system (GIS) data used cannot totally be ruled out. We developed an approach for temporally adjusting NO_2 commute concentrations based on spatial models of annual average air pollutant levels. We computed hourly adjustment factors separately for main and side streets which we applied to legs corresponding to time of the day and road class. With this approach, we took into account differences in daily patterns of traffic volume, NO_2 levels and composition of vehicles at main and side streets. However, classification of the legs by main and side road was not straightforward. There are two GIS road networks available for Switzerland, TeleAtlas and VECTOR25 (Federal Office of Topography swisstopo), both of which were used in our simulation (see article 2, chapter 4). Both networks have a different road classification. We might have misclassified some roads. Furthermore, even small positional errors of roads, buildings and geo-coded home and work addresses have been reported to potentially introduce bias and error in the estimates of exposure to traffic-related air pollution (Zandbergen, 2007).

Finally, the reason for smaller NO_2 contributions of commute to total exposure than for UFP may be also due to differences in spatial and temporal variability of NO_2 and UFP. Recent studies have observed higher contrasts between street sites and urban background for UFP and BC than NO_2 (Boogaard et al., 2011). An investigation on the variation of NO_2 and NO_x concentrations between and within 36 European cities found substantial differences in the street to urban background ratios across study areas. For Basel, a ratio of 1.24 was reported, which is relatively small compared to the overall within city range of 1.09 to 3.16 (Cyrys et al., 2012). Hence, we might have underestimated traffic-related air pollution exposure in traffic by using NO_2.

Nevertheless, despite potential uncertainties of simulated NO_2 concentrations we could show that ignoring time-activity patterns misclassifies exposure and therefore leads to bias in health effect estimates. We also showed that using inhalation dose estimates (i.e. considering transportation mode specific breathing rates) instead of exposure estimates can have different implications in health effects (article 3, chapter 4). Since the NO_2 relative contributions of commute and work/school to total exposure and inhalation dose were rather low compared to those measured in the personal UFP monitoring study, overcoming the modeling limitations would even imply stronger bias in assessments of long-term health effects of traffic-related air pollution. For UFP, the health risk associated to exposure to high UFP concentrations of small particles in traffic may even be increased due to a high potential of particle deposition in the lower airways.

6.5 Strengths and limitations

This doctoral thesis has several strengths. First, it includes several methods of exposure assessment to study commuter exposure to traffic-related air pollution. Individual commuter exposure was studied in various transport microenvironments by means of personal measurements. Additionally, commuter exposure was modeled using actual commuter behavior data of a representative population sample. Second, two important indicator pollutants, UFP and NO_2, of traffic-related air pollution were addressed. With increasing concern that UFP have likely different and independent health effects from larger particles and with the availability of a portable device to measure UFP, the personal monitoring contributes to a better understanding of UFP in transport environments. For the first time, extensive personal measurements of both particle number concentration and average particle size were carried out repeatedly including different times of the day, week and year. Finally, the contribution of commute to total exposure and its potential implications for epidemiological studies were studied for the first time, to best knowledge, based on 24-hour measurements and based on modeling techniques using actual travel route data.

Limitations of the thesis include that the analysis is based on UFP concentration, average particle size and NO_2, providing limited information on other potentially relevant particle metrics for human health such as particle composition and surface area. A limitation of the personal monitoring study is the rather small sample size. The exposure simulation was performed for NO_2 only. At the time of the project, no applicable air pollution models of other air pollutants were available for the study area. But, in principle, the developed simulation is applicable for any traffic-related pollutant. Furthermore, modeled NO_2 exposure estimates rely only on commuter routes and on home and work/school outdoor locations. In-cabin modification, differences in NO2 concentrations between in-transport microenvironments and building-specific indoor-outdoor ratios were not considered. In addition, the inhalation dose estimates in the NO_2 exposure simulation are limited since they were based on literature-derived estimates and not on physiological characteristics of the subjects and actual physical activity measures.

6.6 Scientific implications

Epidemiological studies on long-term health effects of traffic-related air pollution can benefit from incorporating information on people's mobility in the exposure assignment. As it is important to consider exposure differences between and within homes, work places and other microenvironments, it is also important to reflect heterogeneity of lifestyles, habits and time-activity patterns of the study participants (Dons et al., 2011; Rainham et al., 2010; Steinle et al., 2013). Improvements in the precision of exposure assessments may become increasingly important as levels and spatial ranges of air pollutants are decreasing in many countries where clean air polices have improved air quality. It is recommended to carefully evaluate potential benefits of including individuals' time-activity patters in exposure assessment (see Figure 6-1).

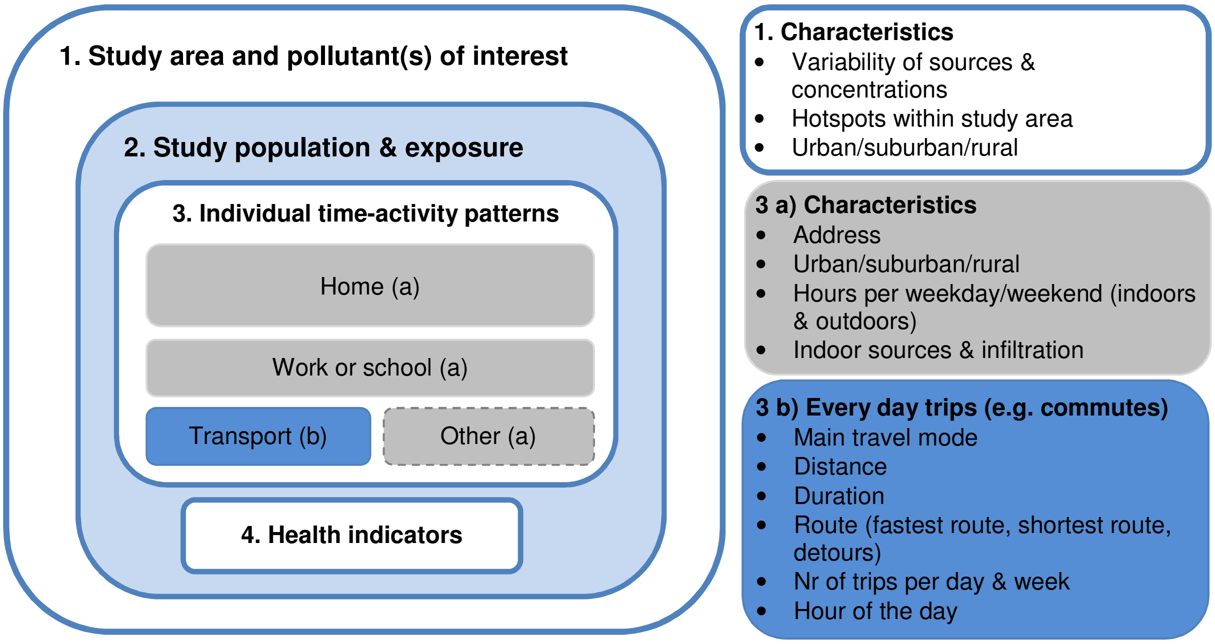

Figure 6-1. Conceptual illustration of air pollution exposure assessment. The boxes on the right include parameters that likely affect exposure and are recommended to be collect from study participants.

First, the study area and the air pollutant and/or source of interest are initial indicators of the expected variability of concentrations. For pollutants with a rather homogenous spatial and temporal spread such as PM_{10}, assessing individual microenvironments might be not appropriate. Second, the question of whether to include commuter behavior and other time-activities in the exposure assessment are related to characteristics of the study population, for example on the spatial spread of the home, work and school locations and the mobility. Third, if potential improvements of reflecting time-activity patterns are expected, information on regular time-activities and habits could be evaluated by means of *activity spaces*. It is recommended to generally differentiate between a) home, b) work/school, c) other locations of regular daily activities and d) travels between the daily activity locations and home. In the study area covering Basel-City and Basel-Country, improvements in exposure assignment associated to including air pollution exposures at work and school locations were achieved for subjects spending >4h per day (work load >=50%) at those places. Ignoring exposures at work was most profound if differences in air pollution concentrations at work were substantially different than at home (e.g. rural versus urban location). According to our NO_2 simulation, commuter behavior is important to be considered in studies including highly mobile population groups commuting between rural and urban areas. *Other* locations were not addressed in this thesis; however, it might be worthwhile in future studies to correct air pollution exposure measures also for other potential important locations around which people usually organize their daily activities.

Incorporating several activity spaces requires a more flexible exposure assessment which applies multiple methods and potentially makes use of novel methods and technology. For example, a more comprehensive approach could include an outdoor air pollution model, an indoor air pollution model (which estimates infiltration of outdoor concentration and indoor air pollution sources) and personal monitoring to introduce a temporal component, verify personal-ambient relationships and refine individual (commuter) exposure estimates.

The study area and the research question also play a role in defining the spatial resolution of the outdoor air pollution model. As shown in article 2 (chapter 4), for estimating in-transport exposure within a city and being interested also in small-scale variability between roads and districts, a model with a high resolution is recommended. For larger scale epidemiological health assessment studies, models with a coarser spatial resolution are likely adequate, especially when the study site covers urban, suburban and rural areas. For the total study area (Basel-City and Basel-Country), the between-subject variability of NO_2 commuter concentrations was sufficiently reflected by the low resolution dispersion model.

6.7 Policy implications

Healthy commuting

The results of the commuter measurements and simulations to traffic-related air pollution give guidance to policy makers in how to improve healthy commuting. In any case, a close collaboration between city planners, policy makers and health experts is essential for the implementation of such transport policies with benefits for health. The following measures could be taken by policy makers to decrease air pollution exposure during commuting:

Exposure to traffic-related air pollution of cyclists and pedestrians can be reduced by choosing a route with low-traffic. Reduced UFP exposures were seen when cyclists avoided main roads, especially during the elevated ambient UFP concentrations in winter and during morning rush hours. Providing a bicycle road network along low-traffic routes in the urban area could lower the exposure of cyclists. Efforts of increasing the distance between cycling lanes and the major road likely also minimize exposure levels. Probable co-benefits of such bicycle infrastructure include a reduced risk of bicycle accidents and less noise exposure (de Nazelle et al., 2011). A risk assessment study within the TAPAS project showed that cycling has greater benefits than risks to health and reduces carbon dioxide emissions (Rojas-Rueda et al., 2011).

From a public health point of view, using public transportation can be supported as people combine it frequently with walking and cycling, thereby enhancing physical activity. UFP concentrations in buses and trams are rather low in Basel. However, as has been shown in this thesis work, persons using public transportation spend a considerable amount of time waiting at public transportation stops. Such locations, as well as the access to those locations, are often exposed to heavy traffic. It is recommended to pay more attention to the placement of public transportation stops in light of recent policy initiatives promoting the use of public transportation. Exposure to traffic-related air pollution at public transportation stops along busy roadways can be significantly reduced by providing shelters that are oriented away from the road (Moore et al., 2012).

UFP policy

Legislation regulating ambient air quality standards and monitoring population exposure to UFP is part of on-going discussion. The lack of exposure-response relationships for UFP and the absence of standard sampling methods are reasons for the non-existent air quality standards for UFP (Heal et al., 2012). As shown in the Basel monitoring study, UFP concentrations are highly variable in transport environments within the city. Therefore, monitoring stations in different urban areas with different road and traffic characteristics are recommended to accurately assess and quantify population exposure to UFP. It is important to monitor UFP continuously at various locations within and between cities. Research on the environmental and health effects of UFP could benefit from such a database. There is a need for epidemiological studies on long-term exposures to UFP (Heal et al., 2012; Hoek et al., 2010). Ambient UFP monitoring will support the development and validation of UFP air pollution models. The data would further be useful for investigations on the sources of various components of UFP and their relationship with other air pollutants and meteorology.

6.8 Outlook

We showed that there are significant differences in UFP concentrations between transport environments as well as between personal UFP and fixed site measurements. The findings indicate that negative bias can be introduced into health effect estimates when air pollution exposures at work/school and during commute are ignored and only outdoor concentrations at home are considered.

Future work will need to more frequently combine modeling approaches with actual personal exposure measurements of pollutants of interest to validate and spatially and temporally refine exposure estimates. The development of such hybrid models is useful to consider the role of exposure variation at the individual level in large cohort studies (Jerrett et al., 2005). For example, the results generated in the UFP personal monitoring campaign can be used to refine individual exposure estimates to UFP, for example in the SAPALDIA study (Swiss study on Air Pollution and Lung Disease in adults) in Basel where UFP models are built and infiltration of UFP to indoor environments and sources of indoor UFP are assessed.

Personal monitoring is still the most accurate method to assess an individual's spatially and temporally resolved exposure to traffic-related air pollution (HEI, 2010). It has not been feasible to carry out personal measurements with representative samples of the study population in large cohort studies on health effects of traffic-related air pollution. However, this might change in the near future. The development of small, portable personal exposure monitoring instruments is a fast evolving field. The new devices are less noisy, lightweight and less bothersome for participants and show constant improvements in precision, accuracy and battery life. The technology is increasingly also taking advantage of devices that people are using every day such as smartphones (de Nazelle et al., 2013; Steinle et al., 2013). Continued improvements are also expected in the development of devices which measure multiple pollutants at the same time. The integration of such devices with real-time tracking of individual time-activity patterns and physical activity measurements are even more promising. In any case, with decreasing prices of small personal monitors, ideally with multiple air pollutants sensors and simultaneously recording location and time, it will be possible to apply them in larger population samples.

To further improve the simulation of commuter exposures to traffic-related air pollution, a more consistent understanding of within transportation mode variability of traffic-related air pollutants and relationships between different traffic-related pollutants is needed. This would help to better estimate population-based in-traffic exposures by means of a few local measurements. In particular, improved knowledge of different vehicle ventilation characteristics on the in-cabin exposure modification is needed to predict commuter exposure based on on-road concentrations.

Future research is warranted to define the population sub-groups for which the assessment of commute exposure and related health effects is most vital. For example, UFP concentration during commute may be of less importance for smokers or people who encounter high occupational UFP concentrations, such as, for example, professional drivers or construction workers. Similarly, such commuter exposure simulations and measurements are also needed in additional cities, especially in the developing world where a lack of in-traffic exposure studies has been identified (Both et al., 2013; Knibbs et al., 2011; Knibbs and Morawska, 2012). Findings from the Basel studies may not be generalizable to other cities of the world.

The focus of this thesis was the measurement and modeling of commuter exposure to traffic-related air pollution. Actual health effects of those commute exposures were not studied. However, a better understanding of health effects of in-transport exposures to air pollution can support health impact assessments of air pollution exposure and transport policies. For instance, only a handful of studies have investigated short-term health effects of in-traffic UFP exposures in real-world settings. Reported effects range from inflammation and signs of lower airway irritation, decreased lung function, oxidative DNA damage to asthma exacerbation (Knibbs et al., 2011). Further research is needed on the short- and also long-term health effects of in-transport exposures. There is a need for studies that investigate the role of short-term exposure and inhalation dose in long-term exposures. Moreover, potential independent health effects of exposure and inhalation dose are not clear. In regard to UFP, efforts are warranted to better differentiate between the effects of UFP and other pollutants and to separate effects of various chemical components of UFP (Knibbs et al., 2011; Rueckerl et al., 2011).

Exposure estimation to traffic-related air pollution and the assessment of related health effects intersects various field of research from health sciences, environmental sciences, psychology, informatics, etc., thus it is a highly multi-disciplinary field. When expanding my literature research to other areas of research not directly related to exposure assessment, I came across several interesting (new) approaches to study time-activity patterns, place and health in transport geography, social sciences and geo-informatics. It is strongly recommended to build and strengthen collaborations between research areas and include expertise from several fields.

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8 APPENDICES

8.1 Supplemental material to Article 1

Appendix A. Details on route of sub-study 2

The annual average traffic intensity on the 2.6 km route (50 km/h speed limit) was 12,430 vehicles/day with 4.5% heavy duty vehicles. The route is served by one bus line and two tram lines. The chosen road segment featured two to four car lanes, two tram tracks in the middle of the street, eight traffic lights at intersections, as well as three bus and nine tram stops. Pedestrian sidewalks were partly separated from the street by a parking lane. Cyclists shared the right-most section of the car lanes. With the absence of any steep slopes, the route is flat with minimum and maximum altitude of 270 and 278 meters above sea level, respectively.

Appendix B. MiniDiSC handling and inlets

Before and after each measurement, zero readings of the miniDiSC were checked with a HEPA inlet and the clock was synchronized with the commuter's watch and a global positioning system (GPS, Wintec WBT-202). The instrument was placed in a backpack and inlets were plac1ed near the breathing zone on the shoulder strap of the backpack. During car measurements, the instrument was put on the front passenger seat with the inlet attached to the headrest.

Inlets and Tubing

A 60 cm long non-conductive Tygon tubing was used as inlet. A few flexible tube inlets (Tygon, conductive antistatic silicone tubing, polyvinyl chloride (PVC)) were tested in co-located measurements before the measurement campaign. A comparison of a PVC and antistatic silocone tubing is provided in Table B. The conductive antistatic tubing seriously affected the instrument performance and maintenance due to contaminating the corona wire charging. Likely the off gassed siloxanes from the tubing deposited on the corona wire and thus affected the charging efficiency especially after long-term use. Furthermore, electrically conductive silicone tubing, which is recommended for aerosol sampling into particle counters, has been shown to contaminate samples by siloxanes (Yu et al., 2009). Being non-conductive, PVC tubing underestimated the particle counts due to particle loss to tube walls and thus both of these tubing were not used further. The correlation between co-located personal measurements with two miniDiSCs with Tygon inlets was high (Spearman corr: 0.998) (Table B).

Table B. Comparison of minute average ultrafine particle number (UFP) concentration and average particle size of two MiniDiSC (MD) devices during personal measurements fitted with polyvinyl chloride (PVC), conductive antistatic (antistatic) and tygon tubing.

		UFP concentration (p/cm³)					Average Particle Size (nm)				
Tubing		MD1	MD2	abs diff (%)	S_{corr}[1]	P_{corr}[2]	MD1	MD2	abs diff (%)	S_{corr}[1]	P_{corr}[2]
PVC & antistatic (3,295 minutes)	mean	6'999	7'768	18.46	0.985	0.987	62.35	59.20	5.42	0.979	0.980
	SD	9'122	9'248	10.35			13.56	11.49	3.17		
	median	3'763	4'444	15.49			60.83	57.70	5.12		
	min	515	893	0.09			17.02	18.38	0.01		
	max	141'352	145'004	124.19			98.55	105.06	27.50		
Tygon (1,629 minutes)	mean	7'125	7'628	7.34	0.998	0.988	46.36	47.05	5.03	0.946	0.928
	SD	9'762	10'948	9.36			8.79	8.55	4.08		
	median	4'538	4'630	3.83			46.66	47.56	4.08		
	min	612	722	0.00			14.50	14.83	0.00		
	max	135'387	163'955	138			77.29	73.80	41.29		

[1]spearman correlation coefficient
[2]pearson correlation coefficient
SD: standard deviation, min: minimum, max: maximum, abs diff: absolute difference

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Appendix C. Weather parameter during measurements

Table C. Description of weather parameters and ambient ultrafine particle number concentration (UFP) during the measurements of sub-studies 1, 2 and 3 (arithmetic mean (standard deviation)).

		n(minutes)	UFP (p/cm^3)	wind speed (m/s)	wind direction (°)	temp (°C)	rel. humidity (%)
Sub-study 1[1]							
winter		505	19'493 (7'148)	2.0 (1.4)	180.1 (87.7)	1.0 (2.7)	82.3 (9.3)
mid season		655	12'212 (7'056)	2.2 (1.8)	201.7 (103.7)	14.6 (3.6)	65.1 (16.0)
summer		658	7'668 (5'932)	1.8 (1.0)	267.7 (56.7)	20.4 (4.5)	62.2 (18.2)
annual		1'818	12'609 (8'161)	2.0 (1.5)	219.5 (92.3)	12.9 (8.7)	68.8 (17.5)
Sub-study 2[1]							
Mode							
Walking		1'272	13'589 (5'147)	1.3 (0.5)	125.6 (96.0)	17.9 (5.6)	50.5 (15.4)
Bicycle		532	13'297 (5'302)	1.3 (0.6)	113.2 (107.9)	19.2 (6.1)	48.2 (15.3)
Bus		392	13'405 (5'383)	1.2 (0.6)	128.4 (115.3)	18.0 (6.3)	49.8 (14.3)
Tram		507	14'260 (6'597)	1.5 (0.5)	125.8 (102.1)	18.2 (5.0)	50.5 (15.0)
Car		586	15'607 (4'393)	1.5 (0.6)	120.2 (85.4)	18.5 (4.7)	57.0 (19.3)
Time period[2]							
rush hour	AM&PM	1'213	17'293 (5'887)	1.5 (0.6)	151.1 (109.3)	15.8 (6.7)	59.6 (21.0)
	AM	642	20'822 (5'192)	1.6 (0.7)	127.7 (10.7)	10.8 (4.0)	77.1 (10.2)
	PM	571	13'122 (3'429)	1.5 (0.5)	178.8 (158.8)	21.6 (4.0)	39.0 (6.7)
non rush hour	AM&PM	1'152	13'831 (5'028)	1.3 (0.5)	120.1 (123.8)	19.2 (4.5)	45.4 (11.3)
	AM	607	16'577 (4'547)	1.1 (0.5)	92.9 (75.4)	17.2 (3.7)	52.8 (9.5)
	PM	545	10'730 (3'550)	1.5 (0.5)	150.0 (157.0)	21.4 (4.4)	37.0 (6.3)
weekend		924	10'845 (2'629)	1.2 (0.6)	90.7 (48.8)	20.1 (3.7)	49.4 (10.9)
total		3'289	14'420 (5'536)	1.4 (0.6)	124.7 (105.9)	18.1 (5.6)	52.0 (16.9)
Sub-study 3[3]							
Winter[4]	total	8	13'873 (5'198)	1.4 (0.7)	171.4 (85.1)	4.9 (4.8)	82.0 (10.1)
	commute[5]	8	21'015 (9'873)	1.2 (0.6)	194.7 (85.9)	4.8 (5.2)	84.6 (8.2)
Spring	total	8	11'860 (5'349)	1.4 (0.9)	158.6 (111.9)	16.5 (6.5)	53.9 (17.1)
	commute[5]	8	15'700 (7'606)	1.4 (1.1)	152.3 (117.8)	15.5 (7.7)	57.2 (20.1)
Summer	total	8	10'453 (5'224)	1.6 (0.8)	209.1 (88.1)	22.4 (6.1)	66.9 (17.9)
	commute[5]	8	13'271 (5'309)	1.7 (0.7)	203.3 (76.5)	21.1 (6.0)	71.5 (17.4)
Total	total	24	11'460 (5'382)	1.5 (0.8)	178.8 (98.3)	14.7 (9.3)	67.5 (19.2)
	commute[5]	24	15'347 (7'380)	1.4 (0.9)	182.2 (97.7)	13.7 (9.3)	70.9 (19.6)

[1]Data source for sub-study 1 and 2: Suburban background station, UFP was measured with a TSI CPC 3775, 10-minute average sampling time, [National Air Pollution Monitoring Network (NABEL), Switzerland].
[2]AM = morning, PM = afternoon
[3]Data source for sub-study 3: Residential fixed site, UFP was measured with a TSI CPC 3022, 30-minute average sampling time
[4]UFP data was available only for three days in winter

Appendix D. Data cleaning and quality control of the miniDiSC data

Raw data was exported and checked with the miniDiSC software (JAVA tool, version 1.191) for potential instrument errors during sampling such as too high/low flow, problems with the current detection or corona voltage. As a preliminary check, unrealistic ultrafine particle (UFP) concentrations smaller than 500 particles/cm³ and/or particle size >250 nm were excluded. Particle sizes smaller 15 nm were flagged as the instruments approaches its measurement limits. In addition, data were checked manually for unreliable measurements and outliers using the filled out time-activity diaries. Time-activity diaries were filled out during each sampling day, collecting information on potential exposure determinants such as passing cigarette smokers, trucks and buses during walking and cycling. Measurements intervals that were contaminated by exposure to second hand smoke were excluded from the analysis of sub-study 1 and 2. In addition, in sub-study 1, two sampling points in the underground bicycle parking garage, one in the shopping centers and one at tram and bus stop had to be excluded due to the influence of other particle sources such as indoor construction and chestnut booths.

Appendix E. Measurement adjustments in sub-study 1

The measurements were adjusted for the temporal variation using data from the suburban background station. Hence, outdoor median UFP concentrations were corrected by computing their ratio to the simultaneously measured UFP at the suburban background station and multiplying this ratio with the daily median UFP concentration at the suburban background station, as shown below (Eq. E):

$$C_{itx} \;/\; C_{back,it} \;*\; C_{back,i} \qquad \textit{(Eq. E)}$$

where:

C_{itx} = median measured UFP concentration on day_i and time $period_t$ in location or $microenvironment_x$
$C_{back,it}$ = median UFP concentration at the suburban background on day_i and time $period_t$
$C_{back,i}$ = median 24-hour UFP concentration at the suburban background on day_i

Table E. Distribution of ratios between the median measured UFP concentrations of sub-study 1 and simultaneously measured median UFP concentrations at the suburban background station.

	n (days)	n (minutes)	mean (SD)	median	min	max
Gundeldingen, *residential urban*	8	227	2.02 (1.86)	0.89	0.95	3.67
Bruderholz, *residential green*	8	241	1.10 (0.91)	0.59	0.45	2.90
Basel city center, *pedestrian*	8	150	2.02 (1.51)	1.54	0.84	5.37
Liestal town center, *traffic*	9	165	2.17 (1.84)	1.59	0.67	6.16
Bus/Tram stop	16	207	2.26 (1.78)	1.35	0.48	6.28

Appendix F. Sub-study 3: UFP concentrations and average particle size during commute and non-commuting

Table F. Average ultrafine particle number concentrations (and standard deviation SD) of individual commute mean and median, and average particle size for two bicycle commuter routes between home and work places (morning AM and afternoon PM) and for non-commuting time by season. Morning commutes include only trips from home to work; evening trips include only trips from work to home (i.e. the commutes from work to home in the mornings are excluded).

					UFP concentration (particles/cm^3)				Particle size (nm)	
			n (trips)	trip duration	average[1]		median[2]		average[3]	
				mean	mean	(SD)	mean	(SD)	mean	(SD)
winter	main street commute	AM	8	14	78'005	(72'859)	58'252	(52'269)	44.0	(8.4)
8 days		PM	4	13	33'912	(14'958)	27'664	(9'511)	49.1	(11.5)
	avoid main street commute	AM	8	15	26'504	(12'748)	22'978	(10'703)	43.7	(6.2)
		PM	4	14	22'939	(9'144)	14'343	(1'816)	63.1	(7.7)
	non-commute				*4'303*	(1'238)	*3'139*	(614)	82.6	(16.9)
spring	main street commute	AM	8	13	32'426	(10'080)	25'711	(5'667)	46.3	(4.3)
8 days		PM	4	10	17'418	(7'838)	10'981	(1'197)	49.6	(7.6)
	avoid main street commute	AM	8	14	33'197	(6'474)	25'787	(4'561)	45.2	(5.0)
		PM	4	13	9'744	(4'241)	7'706	(3'478)	51.5	(6.8)
	non-commute				8'327	(2'419)	6'837	(2'531)	56.6	(6.0)
summer	main street commute	AM	8	12	40'866	(17'891)	26'469	(7'333)	38.8	(3.2)
8 days		PM	4	11	28'220	(16'565)	23167	(13'677)	58.1	(24.1)
	avoid main street commute	AM	8	14	19'317	(4'509)	14'667	(6'385)	54.4	(7.5)
		PM	4	12	16'845	(7'054)	11'164	(4'302)	40.2	(6.7)
	non-commute				8'485	(3'978)	7'087	(2'927)	53.3	(9.0)
total	main street commute	AM	24	13	50'432	(46'400)	36'811	(33'130)	43.0	(6.4)
24 days		PM	12	11	26'340	(10'096)	21'144	(8'767)	47.8	(7.7)
	avoid main street commute	AM	8	15	26'517	(14'271)	20'604	(11'413)	52.3	(15.1)
		PM	4	13	16'510	(8'544)	11'071	(4'154)	51.6	(11.7)
	non-commute				7'038	(3'312)	5'688	(2'841)	64.1	(17.4)

[1]Average UFP concentration during commutes and during non-commuting time
[2]Median UFP concentration during commutes and during non-commuting time
[3]Average particle size diameter during commutes and during non-commuting time

Appendix G. Time-weighted UFP exposures of sub-study 2

Time-weighted median UFP exposures (i.e. median trip concentration × single trip duration) were computed for each trip to compare total UFP exposure among the travel modes.

Table G. Ratios of average time-weighted trip UFP exposures by mode of transport for weekday rush hour. The ratios are expressed in terms of column transport modes divided by row modes.

	Walk	Bicycle	Car	Tram
Bus	5.6	2.1	1.9	1.7
Tram	3.4	1.3	1.1	
Car	2.9	1.1		
Bicycle	2.6			

8.2 Supplemental material to Article 2

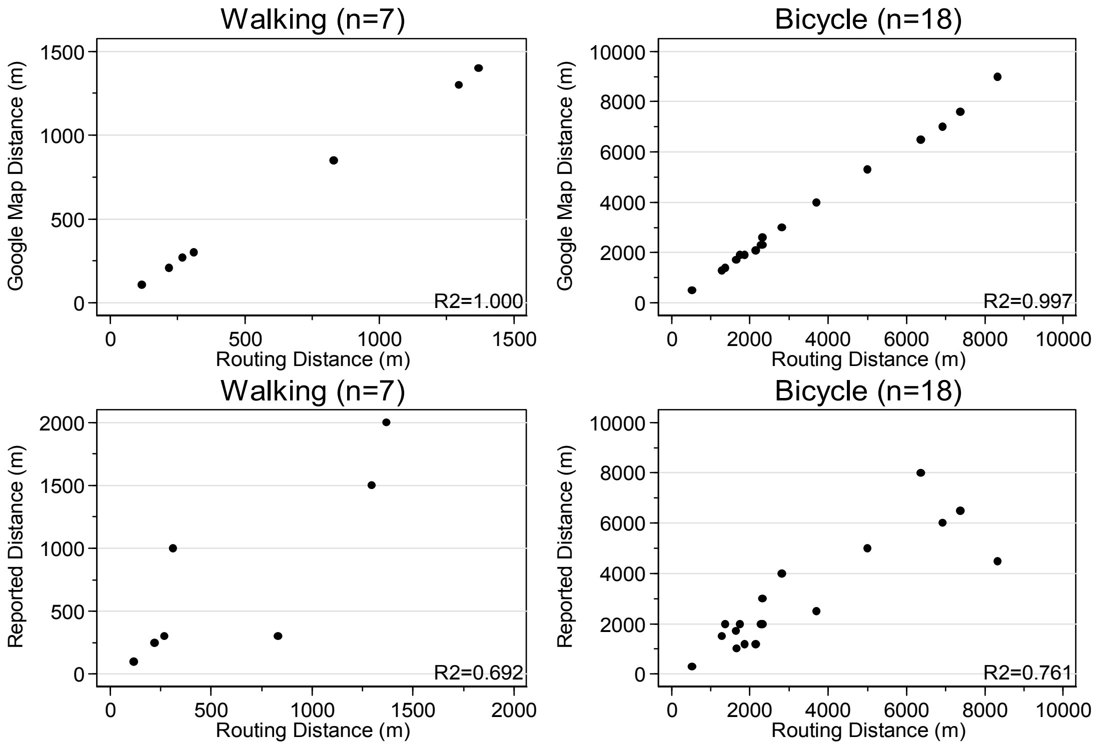
Supplementary Information

Simulation of Population-Based Commuter Exposure to NO_2 Using Different Air Pollution Models

1. Validation of GIS Model

In order to validate the performance of the GIS commuter model used to simulate the walking and bicycle legs in this project, a small validation study was carried out. Information on commuter route and behavior was collected from 36 subjects of the Swiss Tropical and Public Health Institute. First, participants were asked to estimate the duration and distance of their commuter route between home and work locations, as well as to provide information on their commuter mode and route characteristics. Home and work addresses were also collected. Second, participants showed their exact route in Google Map (true distance). Finally, participants' commuter routes between home and work locations were simulated by means of the GIS model developed for this study. Figure S1 shows the comparison between the routing distance (from the GIS model) and the true distance (measured in Googe Map) and reported distance, respectively, for walking and cycling trips. Results are shown for subjects that only walk or cycle to work (*i.e.*, do not combine various travel modes). There is a high agreement between the true distance and the simulated distance (R^2~1.0). Comparisons between the reported distance and the modeled distance are similar to the comparisons with the Microcensus data (Figure S2).

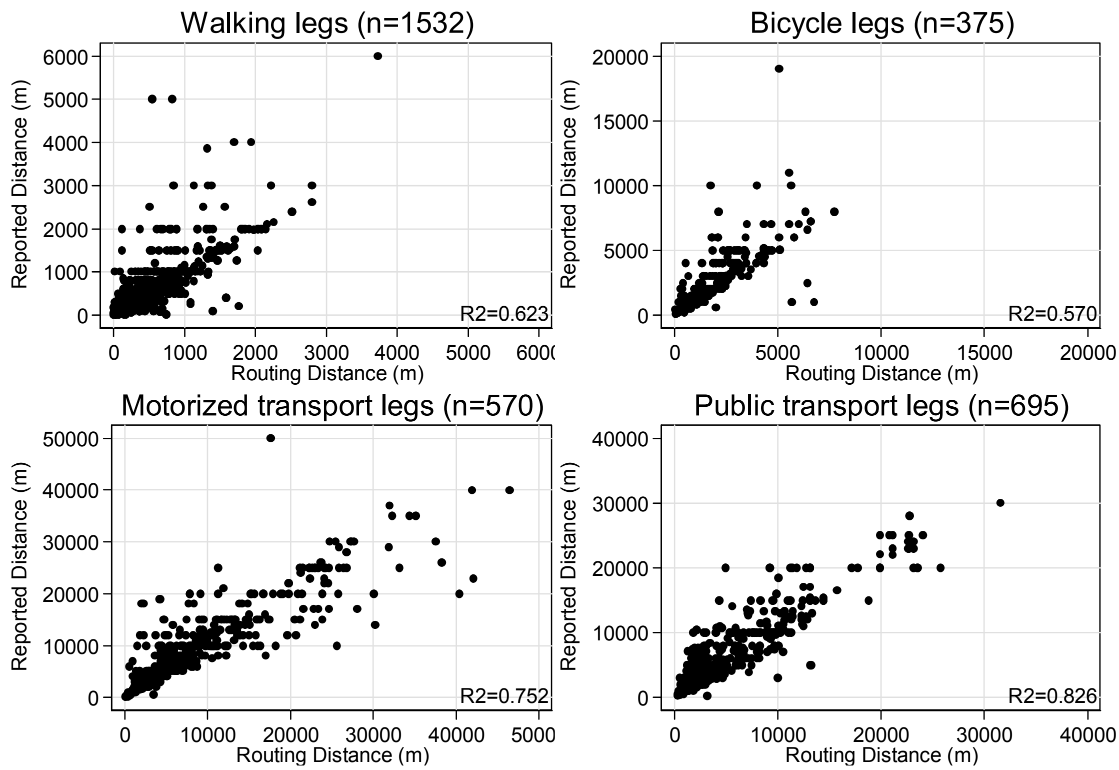
Figure S1. Comparison of simulated routing distance against distance measured in Google map as well as reported distance in a pilot study in Basel.



2. Comparison between the Routing Distance and Reported Distance Provided by the Microcensus Data 2010

Routing distance and reported distance (estimated by the subjects during the interview) were compared (Figure S2) to avoid detours and ensure the plausibility of the legs. Outliers were checked manually and excluded from the analysis if they did not make sense.

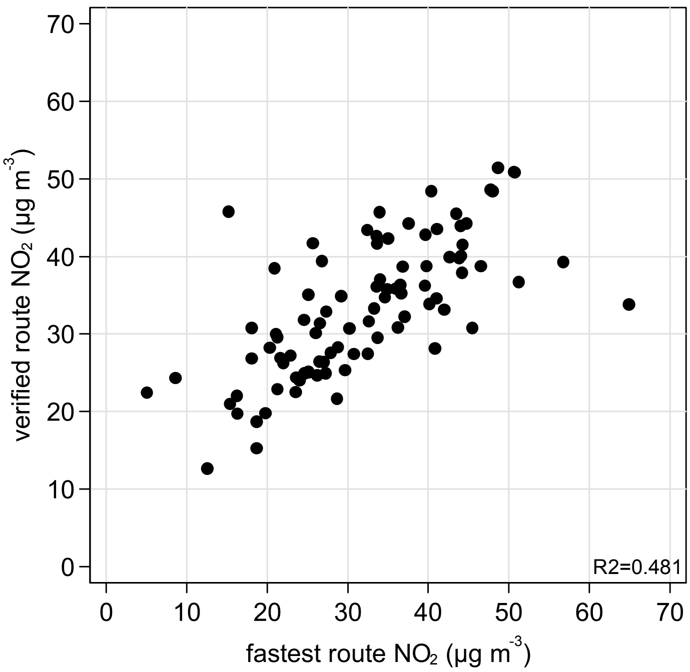
Figure S2. Reported *versus* routing distance. The reported distance was estimated by the subject during the interview.



3. Comparison of Commuter NO_2 Exposure Estimates along Verified and Fastest Car Trips >3km Between Home and Work/School Locations.

We assessed the difference in commute exposure estimates that occur when modeling NO_2 exposure for car trips along the fastest as opposed to the verified route between home and work/school. A subsample of 91 subjects was selected that only commuted by car between home and work/school and had commuter trips longer than 3 km. The verified car trips correspond to the route that was reported during telephone interviews. The fastest route was subsequently simulated based on the same road network (TeleAtlas MultiNet). The PolluMap model was used to estimate commuter exposure to NO_2. Comparing the time-weighted subjects' NO_2 commuter exposure estimates of the two approaches resulted in an R-square of 0.5 (Figure S3). Overall, the mean (±standard deviation) of exposure estimates along the fastest routes (32.2 ± 11.2 μg m^{-3}) was similar to the ones along the verified routes (33.3 ± 8.7 μg m^{-3}). Absolute differences between the corresponding estimates had mean of 5.5 (±6.1) μg m^{-3}.

Figure S3. Subjects' NO_2 commuter exposure (estimated using the PolluMap model) along verified versus fastest car routes between home and work/school locations. Only subjects who commuted by car and had car trips >3 km were included.



4. ESCAPE Basel NO_2 Model (12 April 2012) (Mostly published in Beelen *et al*. 2013, Atmos Env 72(2013) 10–23)

Table S1. Basel NO_2 ESCAPE model: (**a**) describes the model (with VIF—Variable Inflation Factor); (**b**) its performance (with Maximum Cook's distance); (**c**) its leave-one-out cross-validation; and (**d**) its Moran's I.

		(a) The Model			
	Estimate	Std. Error	t value	Pr(>|t|)	VIF
(Intercept)	5.435E+01	3.306E+01	1.64	0.109	
INTMAJORINVDIST	1.227E−02	2.244E−03	5.47	0.000	1.082
RES5000_500	5.330E−07	2.452E−07	2.17	0.037	1.482
SQRALT	−3.956E+00	1.523E+00	−2.60	0.014	1.379
RES500	1.878E−05	1.063E−05	1.77	0.086	1.181
		(b) Performance			
	R2	Adj_R2	RMSE	MaxCooksD	
Basel ESCAPE model	0.67	0.63	4.48	0.16	
		(c) Cross Validation			
	CV_R2	CV_Adj_R2	CV_RMSE		
Basel ESCAPE model	0.58	0.57	4.83		
(**d**) Moran's I					
	observed	expected	sd	*p* value	
Moran's I	−0.05	−0.03	0.03	0.45	

Predictor Definitions:

INTMAJORINVDIST:	Product of traffic intensity on nearest major road (INTMAJOR) and inverse of distance to the nearest major road (INVDIST). [Veh/day/m]
RES5000_500:	Residential land in a donut with outer radius of 5,000m and inner radius of 500 m. [m^2]
SQRALT:	Square root of altitude. [$m^{1/2}$]
RES500:	Residential land in a circle of radius 500 m. [m^2]

5. Temporal Adjustment

We calculated temporal adjustment factors for each hour of the day separately for main roads and side streets to consider the diurnal pattern of NO_2 levels and differences in hourly traffic volume and composition of vehicles (*i.e.*, with separate counts for personal cars and trucks). Ratios were computed between the annual weekday hourly means and the annual mean concentration measured at the monitoring stations (see Figure S4). While the ratios at the side road were peaked during rush hours, those at the street site were highest during working hours (9–11am; 3–5pm), most likely due to increased truck traffic. Ratios were applied to each commuter leg concentration based on the road classification and start hour of the leg. Table S2 shows the percentage of legs defined as main and side roads separately for study area. Summary statistics of the ratios are provided in Table S3.

Figure S4. Number of commuter legs per hour of the day and annual weekday hourly NO_2 means (±standard deviation) at the two fixed stations (main road and side road) used to compute temporal adjustment ratios. The straight lines represent the annual average NO_2 measured at the two fixed stations.

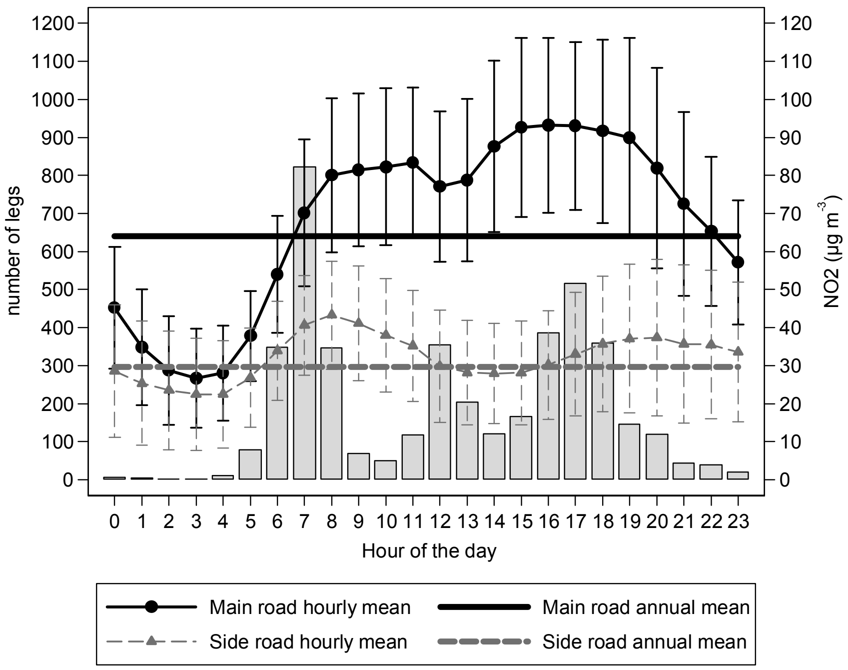


Table S2. Street class applied to legs by travel mode and study area.

Travel Mode	Study Area	N (Legs)	Main Road (%)	Side Road (%)
walking	Basel-City	966	22.6	77.4
	Total area	2583	23.3	76.7
bicycle	Basel-City	204	34.8	65.2
	Total area	385	29.9	70.1
motorized transport	Basel-City	58	51.7	48.3
	Total area	602	79.7	20.3
public transport	Basel-City	259	60.6	39.4
	Total area	733	56.9	43.1
other	Basel-City	18	0.0	100.0
	Total area	37	0.0	100.0

Table S3. Summary of ratios applied to legs and waiting points by study area and road class.

Study Area	Variable	n (Legs)	Mean	(sd)	Median	Min	Max
Basel-City	main road	476	1.25	(0.18)	1.25	0.59	1.45
	side road	1029	1.17	(0.18)	1.14	0.76	1.46
Total Area	main road	1615	1.24	(0.22)	1.25	0.44	1.45
	side road	2725	1.18	(0.17)	1.14	0.76	1.46

6. Comparison of the Three NO_2 Models with Measurements

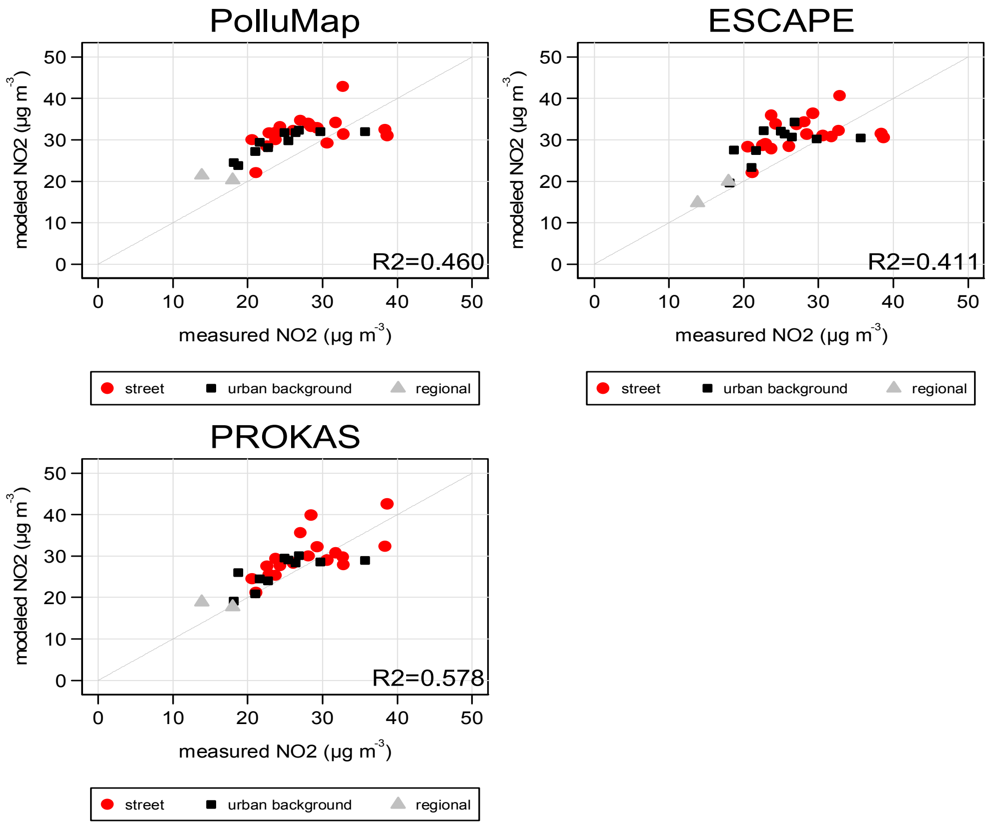
We compared the three models—PROKAS, ESCAPE and PolluMap—to NO_2 measurements from a total of 31 sites within Basel-City from the Swiss study on Air Pollution and Lung and Heart Diseases in Adults (SAPALDIA) (Table S4, Figure S5). These measurements were conducted outside subjects' homes in three biweekly integrated sampling campaigns in 2011 using Passam passive diffusion samplers (Passam AG, Schellenstrasse, Männedorf, Switzerland). To keep the evaluation spatially comparable, we compared the average NO_2 concentrations of each site to the value estimated for the grid value of the three models. Thus, the data do not reflect a proper validation of the models.

Table S4. Comparison between modeled and measured [a] NO_2 (in µg m^{-3})

Model	Street Sites (*n* = 18)			Urban Background Site (*n* = 11)			All Sites [c] (*n* = 31)		
	R^2	Bias [b]		R^2	Bias [b]		R^2	Bias [b]	
		Mean	(sd)		Mean	(sd)		Mean	(sd)
PROKAS	0.44	−2.1	(4.4)	0.50	−1.6	(3.6)	0.58	−1.9	(4.0)
ESCAPE	0.15	−3.6	(5.4)	0.34	−4.4	(4.4)	0.41	−3.7	(4.8)
PolluMap	0.17	−4.1	(5.2)	0.67	−4.7	(3.1)	0.46	−4.3	(4.4)

Note: [a] Measurements were conducted during three seasons (two-week samples) in 2011 as part of the Swiss SAPALDIA study (Basel-City only); [b] The bias is calculated as the difference (predicted-measured) NO_2, shown as mean and standard deviation (sd); [c] Two sites are classified as regional background sites.

Figure S5. Comparison of NO_2 models with measurements of 31 sites from the 2011 SAPALDIA study.



7. Study Population

Table S5. Characteristics of the study population.

		Basel-City		Total Area	
subjects	n	258		736	
female	%	59.3		50.3	
age	mean (sd)	35.0	(17.0)	36.7	(17.6)
work fulltime (≥90%)	%	51.2		53.8	
work ≥50%–89%	%	16.3		16.4	
work <50%	%	7.0		7.6	
in education	%	7.4		5.7	
<15 years old	%	18.2		16.4	
subjects with 2 commuter trips/day [a]	%	84.1		84.4	

Note: [a] The rest of the subjects had four trips per day.

8. Spearman Correlation Coefficients between NO_2 Models

Table S6. Spearman correlation coefficients of daily NO_2 commuter estimates between the three air pollution models in the study area Basel-City.

Model Comparison				
	n	PROKAS-ESCAPE	PROKAS-PolluMap	ESCAPE-PolluMap
concentration (legs)	1175	0.89	0.85	0.91
commuter concentration [a] (subject)	258	0.87	0.81	0.86
commuter exposure [b] (subject)	258	0.99	0.99	0.99
commuter dose (subject)	258	0.99	0.99	0.99

Note: [a] Sum of leg concentrations divided by the number of legs; [b] Concentrations multiplied by durations, includes waiting time between legs. All correlations are significant ($p < 0.05$).

8.3 Supplemental material to Article 3

1. Waiting time and main travel modes

The subjects' main commuter travel mode is defined as the mode used for the longest distance during daily commutes. Table S1 summarizes the duration of each travel mode (contributing travel mode) and the waiting time for each subject's main commuter mode. The subjects who use public transportation as their main travel mode spent a considerable amount of their commute time waiting. In fact, the subjects who used public transportation as their main commuter mode spent 17.6 ±11.78% of their commute time on average waiting at public transportation stops.

Supplementary Information, Table S1. Commute duration (in minutes) per day in the total study sample (690 subjects) according to travel mode and each subject's main mode.

			Duration (minutes)			
main travel mode	**contributing travel mode**	***n* (subjects)**	**mean ±SD**	**median**	**min**	**max**
walk	walk	129	30 ±21	25	2	120
	waiting	10	10 ±9	6.5	1	28
	bicycle	3	14 ±10	10	7	25
	motorized transportation	2	3 ±2	2.5	1	4
	public transportation	11	13 ±7	13	4	26
	other	1	3 .	3	3	3
bicycle	walk	10	13 ±10	10	2	30
	waiting	7	8 ±11	2	1	30
	bicycle	121	30 ±18	30	4	125
	motorized transportation	1	5 .	5	5	5
	public transportation	0	. .	.	.	.
	other	0	. .	.	.	.
motorized transportation	walk	56	11 ±15	7	1	90
	waiting	34	9 ±16	3.5	1	84
	bicycle	5	23 ±16	18	4	40
	motorized transportation	220	37 ±21	35	4	134
	public transportation	9	19 ±14	14	3	53
	other	0	. .	.	.	.
other	walk	1	14 .	14	14	14
	waiting	2	6 ±1	5.5	5	6
	bicycle	0	. .	.	.	.
	motorized transportation	0	. .	.	.	.
	public transportation	2	27 ±9	26.5	20	33
	other	12	24 ±13	20	6	49
public transportation	walk	196	23 ±14	20	2	68
	waiting	190	15 ±13	12	1	70
	bicycle	6	26 ±17	28	7	50
	motorized transportation	11	16 ±10	12	4	30
	public transportation	198	40 ±21	38	4	121
	other	1	28 .	28	28	28

min: minimum, max: maximum

2. Variance and correlation coefficients of NO_2 concentrations

Supplementary Information, Table S2. Variance of NO_2 concentration (in µg m^{-3}).

microenvironment	subjects (n)	variance
outdoors at home	680	39.8
outdoors at work or school	680	34.5
commute	680	57.5

Supplementary Information, Table S3. Pearson correlations between microenvironments.

microenvironments		total sample	within Basel-City	within Basel-Country	between Cantons
H	W	0.49	0.22	0.47	-0.46
H	C	0.69	0.44	0.62	0.29
W	C	0.72	0.39	0.65	0.12[a]

H: outdoors at home; W: outdoors at work/school, C: commute
[a] p>0.05

Supplementary Information, Table S4. Spearman correlations between microenvironments.

microenvironments		total sample	within Basel-City	within Basel-Country	between Cantons
H	W	0.45	0.15	0.55	-0.50
H	C	0.68	0.46	0.64	0.31
W	C	0.66	0.22	0.69	0.07[a]

H: outdoors at home; W: outdoors at work/school, C: commute
[a] p>0.05

3. Summary statistics of NO_2 exposure and dose for sub-samples

Supplementary Information, Table S5. Summary statistics for population NO_2 exposure (time-weighted over a 7-day week, µg m-³) and weekly inhaled dose (µg m-³ × minutes × ventilation ratio) for sub-samples. NO_2 estimates are shown for outdoors at home (H), time-weighted home and work/school (HW) and time-weighted home, work/school and commuting (HWC).

NO_2 estimate	mean ±SD	min	p5	p25	median	p75	p95	max	iqr	ratio p95/p5
subjects commuting within Basel-City (240 subjects)										
H_{exp}	30.9 ±3.6	19.0	23.3	29.8	31.7	33.1	34.6	51.1	3.3	1.48
$H_{dose}{}^{a}$	311.3 ±36.4	191.2	234.9	300.2	319.4	333.2	348.4	515.0	32.9	1.48
$HW_{exp}{}^{b}$	31.1 ±3.1	20.1	24.7	29.9	31.8	32.9	34.3	48.8	3.0	1.39
$HW_{dose}{}^{a,\,b}$	313.2 ±31.1	203.0	248.8	301.6	320.1	331.9	345.3	492.0	30.2	1.39
HWC_{exp}	31.2 ±3.0	20.3	24.8	30.0	31.9	33.1	34.4	48.6	3.1	1.39
$HWC_{dose\ moderate}{}^{a,\,c}$	319.2 ±31.3	208.2	251.0	307.0	326.4	337.7	351.0	498.6	30.7	1.40
$HWC_{dose\ high}{}^{a,\,d}$	325.4 ±33.5	221.8	252.7	311.0	331.6	342.2	365.9	529.1	31.1	1.45
subjects commuting within Basel-Country (270 subjects)										
H_{exp}	21.0 ±4.7	11.5	13.7	17.6	21.1	24.1	28.7	41.1	6.5	2.09
$H_{dose}{}^{a}$	211.8 ±47.0	116.4	137.6	177.0	212.3	242.4	289.3	414.7	65.5	2.10
$HW_{exp}{}^{b}$	21.4 ±4.2	11.5	14.8	18.3	21.6	24.0	28.1	37.7	5.7	1.90
$HW_{dose}{}^{a,\,b}$	215.6 ±42.6	116.1	149.4	184.6	217.7	242.4	283.0	379.7	57.8	1.89
HWC_{exp}	21.6 ±4.2	11.5	14.9	18.5	21.8	24.2	28.5	37.8	5.7	1.91
$HWC_{dose\ moderate}{}^{a,\,c}$	219.3 43.0	116.2	151.6	187.5	220.8	247.8	288.1	381.4	60.4	1.90
$HWC_{dose\ high}{}^{a,\,d}$	221.0 43.9	116.2	151.6	187.5	225.4	248.7	295.5	381.4	61.2	1.95
subjects commuting between Basel-City and Basel-Country (170 subjects)										
H_{exp}	25.4 ±5.8	12.4	16.4	21.5	25.2	29.6	34.3	54.5	8.1	2.09
$H_{dose}{}^{a}$	256.2 ±58.2	125.5	164.9	216.3	253.7	298.1	345.9	549.8	81.7	2.10
$HW_{exp}{}^{b}$	26.6 ±4.0	17.5	20.0	24.1	26.6	29.4	32.6	49.2	5.3	1.63
$HW_{dose}{}^{a,\,b}$	267.7 ±40.5	176.7	201.5	242.9	267.7	295.9	328.9	495.6	53.0	1.63
HWC_{exp}	26.9 ±3.9	18.2	20.7	24.8	26.9	29.6	32.6	48.8	4.7	1.57
$HWC_{dose\ moderate}{}^{a,\,c}$	275.1 39.2	189.1	210.3	253.5	274.5	301.9	332.1	499.7	48.4	1.58
$HWC_{dose\ high}{}^{a,\,d}$	278.6 42.9	189.1	210.3	253.6	275.6	303.2	345.1	526.8	49.6	1.64

SD: standard deviation, min: minimum, p: percentile, max: maximum, iqr: interquartile range

[a] Inhalation dose estimates are shown in 1000 µg m-³

[b]Average commute exposure is assumed to be equal to the average outdoor exposure at home

[c]Ventilation ratios applied: motorized transport: 1; public transport: 1; walking: 1.7; bicycle: 2; home and work location: 1.

[d]Ventilation ratios applied: motorized transport: 1; public transport: 1; walking: 1.7; bicycle: 5.6; home and work location: 1.

4. Contribution of commute to total NO_2 exposure and dose by sub-group

Supplementary Information, Table S6. Contribution of home, work and commute (in %) to the total weekly NO_2 exposure and inhalation dose adjusted NO_2 exposure for the total sample and subgroups.

Sample	Estimate	Micro-environment	n (subjects)	mean±SD	median	min	max
total population	exposure	home	680	73.4±7.4	73.9	37.8	89.8
		work/school	680	23.4±6.4	23.5	8.4	48.8
		commute	680	3.2±2.3	2.7	0.1	13.5
	dose moderate	home	680	72.6±7.5	73.3	36.1	88.3
		work/school	680	23.1±6.4	23.4	8.3	47.7
		commute	680	4.3±3.0	3.6	0.2	17.3
	dose high	home	680	71.7±7.6	72.6	36.1	87.1
		work/school	680	22.9±6.4	22.8	7.8	47.7
		commute	680	5.4±4.2	4.3	0.2	33.0
cummuting within Basel-City	exposure	home	240	75.4±5.0	74.6	60.9	89.8
		work/school	240	22.0±4.8	23.2	8.4	34.5
		commute	240	2.6±1.6	2.2	0.2	9.6
	dose moderate	home	240	74.4±5.0	73.9	60.2	88.3
		work/school	240	21.7±4.8	23.0	8.3	34.5
		commute	240	3.9±2.2	3.5	0.4	14.3
	dose high	home	240	73.1±5.5	73.0	57.2	86.5
		work/school	240	21.3±4.8	22.2	7.8	34.5
		commute	240	5.6±3.9	4.6	0.9	27.4
cummuting within Basel-Country	exposure	home	270	74.7±7.1	74.6	37.8	87.2
		work/school	270	22.5±6.2	22.8	10.7	48.8
		commute	270	2.8±2.3	2.1	0.1	13.5
	dose moderate	home	270	74.1±7.2	73.9	36.1	87.1
		work/school	270	22.3±6.2	22.5	10.7	47.7
		commute	270	3.7±3.0	2.7	0.2	17.3
	dose high	home	270	73.5±7.3	73.6	36.1	87.1
		work/school	270	22.1±6.2	22.2	10.7	47.7
		commute	270	4.3±3.8	3.1	0.2	24.4
cummuting between Basel-City and Basel-Country	exposure	home	170	68.5±8.5	68.0	47.2	86.9
		work/school	170	26.8±7.5	27.9	10.0	44.9
		commute	170	4.7±2.4	4.2	0.7	12.9
	dose moderate	home	170	67.6±8.6	67.5	45.9	86.9
		work/school	170	26.4±7.4	27.6	9.8	43.7
		commute	170	6.0±3.2	5.4	0.7	16.6
	dose high	home	170	66.9±8.6	67.0	40.9	86.9
		work/school	170	26.2±7.5	27.5	9.8	43.7
		commute	170	6.9±4.5	5.9	0.7	33.0

SD: standard deviation, min: minimum, max: maximum

5. Estimated bias factors in sub-groups

Supplementary Information, Table S7. Estimated bias factors and 95% confidence intervals in the health effect estimates by scenario for the subjects commuting within Basel-City, within Basel-Country, and between Basel-City and Basel-Country.

Scenario	estimate used	assumed correct estimate	within Basel-City (*n*=240)	within Basel-Country (*n*=270)	between Basel-City & Basel-Country (*n*=170)
1	H_{exp}	HW_{exp}	0.83 (0.80, 0.87)	0.88 (0.85, 0.91)	0.67 (0.63, 0.71)
2	H_{exp}	HWC_{exp}	0.82 (0.79, 0.86)	0.87 (0.84, 0.91)	0.64 (0.60, 0.67)
3	H_{dose}[a]	$HWC_{dose\ moderate}$[b]	0.83 (0.80, 0.87)	0.88 (0.84, 0.92)	0.64 (0.59, 0.68)
4	H_{dose}[a]	$HWC_{dose\ high}$[c]	0.83 (0.76, 0.89)	0.89 (0.85, 0.93)	0.68 (0.61, 0.73)
5	HW_{exp}	HWC_{exp}	0.99 (0.98, 0.99)	1.00 (0.99, 1.00)	0.96 (0.95, 0.96)
6	HW_{dose}[a]	$HWC_{dose\ moderate}$[b]	1.00 (0.98, 1.02)	1.00 (1.00, 1.01)	0.96 (0.94, 0.98)
7	HW_{dose}[a]	$HWC_{dose\ high}$[c]	0.99 (0.92, 1.05)	1.02 (1.00, 1.03)	1.01 (0.95, 1.06)

H: outdoors at home; HW: home and work, HWC: home, work/school and commuting

[a]Ventilation ratios applied: home location: 1, work location: 1

[b]Ventilation ratios applied: motorized transport: 1; public transport: 1; walking: 1.7; bicycle: 2; home and work location: 1.

[c]Ventilation ratios applied: motorized transport: 1; public transport: 1; walking: 1.7; bicycle: 5.6; home and work location: 1.