Pilot Study for the Development of a Personal Exposure Model for Personal Ultrafine Particle Exposure

Inaugural-Dissertation
zur
Erlangung des Doktorgrades
Dr. rer. nat.

der Fakultät für
Biologie

an der

Universität Duisburg-Essen

vorgelegt von
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aus Dinslaken
Datum der Abgabe: September 2010

Die der vorliegenden Arbeit zugrunde liegenden Experimente wurden am Institut für Medizinische Informatik, Biometrie und Epidemiologie der Universität Duisburg-Essen oder an einer anderen gleichwertigen Einrichtung durchgeführt.

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Tag der mündlichen Prüfung: 03.02.2011

Contents

Summary of Tables

Summary of Figures

Abbreviations

AW	At-work
CB	Contribution
CPC	Condensation Particle Counter
CV	Coefficient of Variation
E	Exposure
EPA	Environmental Protection Agency
EQ	Equation
ETS	Environmental Tobacco Smoke
EURAD	Europäisches Ausbreitungs- und Depositionsmodell
EXPOLIS	European Exposure Assessment Project
GIS	Geographic Information System
HEPA	High Efficiency Particulate Air Filter
IME	Integrated Meteorological-Emission
INF	Infiltration Factor
IT	In-transit
IUTA	Institute of Energy and Environmental Technology
ME	Microenvironment
MS	Microsoft
Ø	Diameter
OP	Other places
PD	Particle Diameter
PM	Particulate Matter
PVC	Polyvinyl Chloride
Q1	1st quartile
Q3	3rd quartile
RI	Residential-indoor
RO	Residential-outdoor
SD	Standard Deviation
UFP	Ultrafine Particles
VOC	Volatile Organic Compounds
WHO	World Health Organisation

Summary

Ultrafine particles (UFP) have been identified as having adverse effects on human health. It is, therefore, important to assess exposure to specific UFP-emitting sources. Assessment of UFP concentrations are very time and cost intensive in large study populations. Modelling is, therefore, a convenient tool for estimating UFP concentrations. To date, models have focussed on ambient air concentration without taking local sources within the breathing space of the participant or individual habits into account.

In order to fill this gap, this study incorporated a population of 22 individuals equipped with a UFP sampler for a period of 48 hours so as to assess personal exposure to UFP and to identify major UFP-emitting sources. With the help of a diary and questionnaire, the UFP data collected could be allocated to specific sources. Furthermore, information from the questionnaire was used in order to develop a personal UFP exposure model which included local and ambient UFP-emitting sources.

This study was able to identify differences in UFP concentrations for specific UFP-emitting sources. Moreover, it was able to identify major contributors to UFP, namely cooking and travelling by car. The data collected in this study, with an overall availability of 912 hours, matches values found in the literature. With respect to UFP modelling, it is seen that the data collected was highly variable for the same UFP-emitting source within an individual and within the entire study population. Moreover, a theoretical model was developed for the assessment of personal UFP exposure. Due to high variability of collected data and the small study population, a validation of this model could not be conducted. This study showed that a higher level of detail is necessary in order to complete the personal UFP model and to perform a validation. This means, that more information on cooking and cleaning methods, such as, boiling, frying, dusting or vacuuming, is needed in order to allocate UFP concentrations to specific exposure situations.

1. Introduction

1.1. Total Suspended Particulates

Total suspended particulates (TSP) are a mixture of solid, liquid, or both solid and liquid particles suspended in the air, commonly known as soot, smoke or dust. These particles vary in size, composition and origin and are conveniently classified according to their aerodynamic properties due to the following reasons:
(a) these properties govern the transportation and removal of particles in the air;
(b) they also govern their deposition within the respiratory system and
(c) they are associated with the chemical composition and sources of particles (WHO, 2003).

Particulate matter (PM) is part of the TSP and can be divided into three fractions according to size:
(a) coarse (< 10µm),
(b) fine (< 2.5µm) and
(c) ultrafine (<0.1µm),
or, alternatively, into two fractions according to their deposition within the human body:
(a) thoracic including particulate matter with an aerodynamic diameter up to 10µm (PM_{10}) which mainly deposit in the upper airways, and
(b) respirable, including PM with an aerodynamic diameter up to 2.5µm ($PM_{2.5}$ and smaller), which are the main particles reaching the alveoli.

Fig. 1.1 shows the various solid particles suspended in air while Tab. 1.1 summarises the different fractions of particulate matter described above.

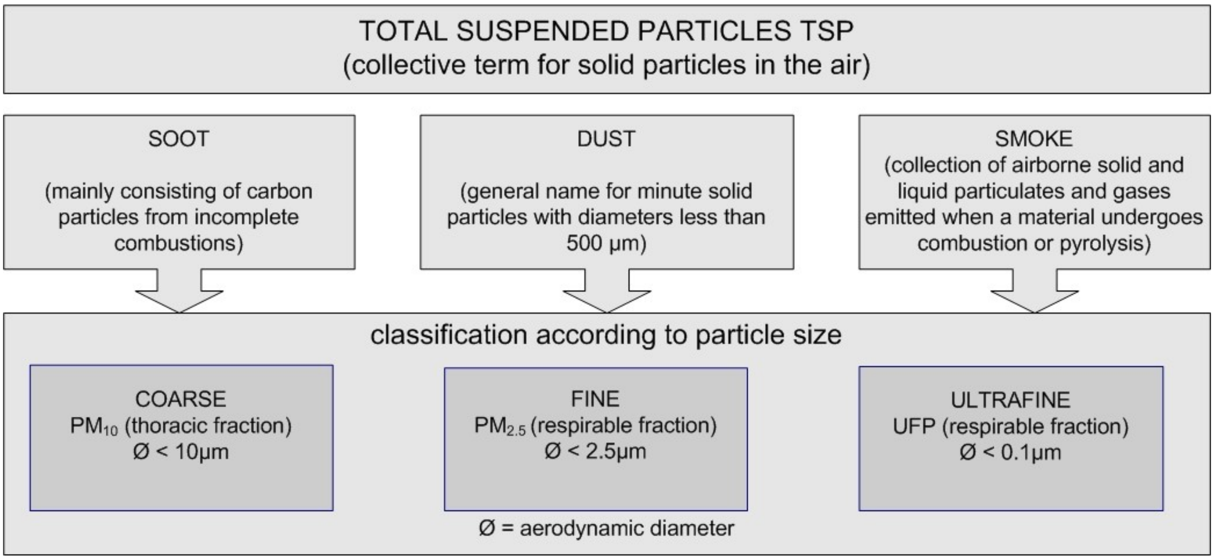

Fig. 1.1 Summary of Total Suspended Particles in the air and their classification according to particle size (Modified after Wikipedia (2005))

This section will now go on to present the various PM fractions according to size.

The Coarse Fraction

Coarse particles (PM_{10}) are mainly derived from natural sources, namely suspension or resuspension of windborne dusts, sea spray, soil, or other crustial materials from roads, farms, mining, windstorms or volcanoes and the combustion of fuel. $PM_{2.5\text{-}10}$ may also consist of pollen, mould, spores and other plant materials (Sioutas et al., 2005; Diapouli et al., 2008; Guo et al., 2008; Koulouri et al., 2008; Maraziotis et al., 2008; Polymeneas and Pilinis, 2008; Yang et al., 2008). Geller et al. (2002) suggested that a substantial portion of coarse particles is also generated from indoor sources and activities, including dusting, cleaning, washing and resuspension. Due to the particles' large size and resulting higher gravitational settling velocity relative to other suspended particles', they can remain suspended in air for several hours (Sioutas et al., 2005). The coarse fraction also includes particles from the fine and UFP fraction.

The Fine Fraction

Particles with an aerodynamic diameter of 0.1 to 2.5µm constitute the fine fraction ($PM_{2.5}$). Most of the acidity (hydrogen ions) in the air and particles with mutagenic activity can be found in this fraction. The fine fraction is mainly formed by combustion of fossil energy sources, such as, oil and its by-products or coal, and mainly consists of organic carbon, elemental carbon and several trace metals originating from combustion processes. Motor vehicles have, consequently, been identified as a primary direct emission source of fine and ultrafine particles liberated into the atmosphere in urban areas (Hitchins et al., 2000; Zhu et al., 2002). Most of the aerosol mass in this category is, therefore, secondary in nature. This means that these particles have formed from reactions of gaseous species, such as sulphur dioxide, nitrogen dioxide and volatile organic compounds (VOC) with atmospheric oxidants by either forming new particles or adding mass to existing particles (Nadadur et al., 2007).

Natural formation of $PM_{2.5}$ is similar to that of the coarse fraction mentioned above. Further ambient sources of organic $PM_{2.5}$ include wood smoke, dust from paved roads, and gaseous precursors of organic aerosol. Major indoor sources identified for $PM_{2.5}$ are namely cooking, cleaning, smoking, various human activities, such as, simple movement, and processes involving burning (Chao and Cheng, 2002). Particles from the fine fraction may remain suspended within the atmosphere for several days and may also be transported over long distances (Sioutas et al., 2005). The fine fraction also includes particles from the UFP fraction.

The Ultrafine Fraction

Ultrafine particles (UFP) are those having an aerodynamic diameter of less than 0.1µm (WHO, 2003). Although UFP dominate ambient air particles by number, they rarely account for more than a small percentage of the total mass (Pekkanen and Kulmala, 2004).

Several processes are known to form UFP in the atmosphere by direct emission, namely combustion originating from traffic or industrial processes which directly emit UFP into the environment. These combustion processes may also emit hot supersaturated vapours, which may experience nucleation and condensation while cooling to ambient temperature. Chemical reactions in the atmosphere may also lead to the formation of chemical species with low vapour pressure (Kittelson, 1998 in; Sioutas et al., 2005).

Additionally, it has been shown that photochemical atmospheric reactions lead to the formation of low-volatility species at ambient temperature. These chemical species may form UFPs while undergoing different nucleation processes (Kulmala et al., 2004; Stanier et al., 2004). Nucleation may also occur on ions and there is strong evidence to suggest that sulphuric acid vapour may also play a role in this process. Ammonia and water vapour are also involved and have been implicated in UFP formation by this method. Other trace gases in the atmosphere, such as, organic compounds, may either participate in the nucleation process or react in the atmosphere to form compounds that nucleate. Due to their extremely low concentrations, the identity and concentrations of these gases are not yet known (Kulmala et al., 2004). The UFP produced by the various methods mentioned are chemically unstable and may revert to their original species (Fig. 1.2).

Several indoor UFP-emitting sources have also been identified, including cooking, cleaning and smoking (Abt et al., 2000). A study focusing on indoor sources of fine and ultrafine particle emissions identified cigarettes, heaters, frying of meat, vacuum cleaners, burning candles and air-freshener sprays as potential sources (Afshari et al., 2005). Furthermore, laser printers and copiers can also be the predominant contributors to ambient UFP concentration in offices (Schripp et al., 2008). The formation and distribution of the three types of particulate matter suspended in air are demonstrated in Fig. 1.2.

Due to their agglomerate structure, UFPs have a larger surface area than spherical particles and can, therefore, carry large amounts of absorbed or condensed toxic air pollutants which can potentially have significant impact on health (Oberdörster, 2001).

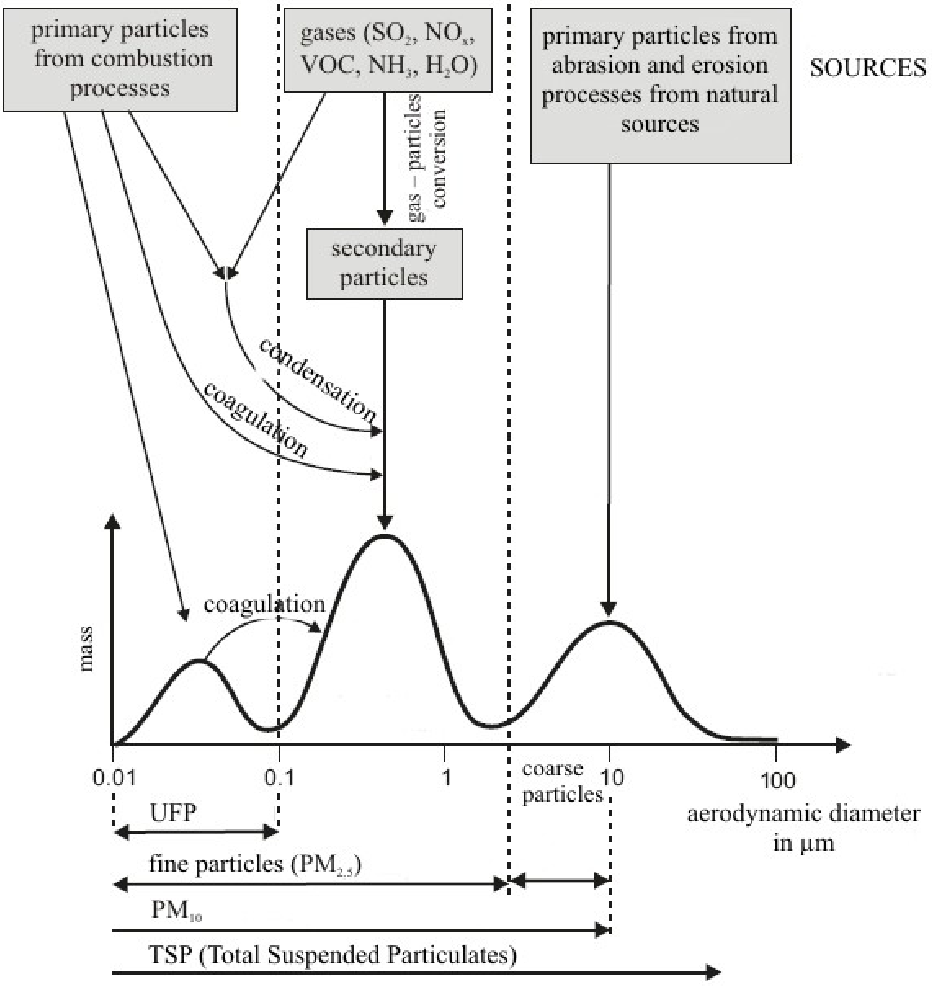

Fig. 1.2 Schematic description of particle formation processes and particle size distribution in ambient air (Modified after EKL ((2007)).

1.2. Deposition of PM

Particle size and number as well as correlated parameters, such as, surface area and density, have been key issues in understanding their physiological effects. Particle size, for example, determines their deposition in the lungs as well as their elimination from the body. Coarse PM, referred to as the thoracic fraction, may reach the larger airways as well as, to a lesser extent, the smallest airways and alveoli. It is mainly the fine and ultrafine particles, known as the respirable fraction that deposit in the alveoli. Out of these two fractions, the ultrafine particles have a substantially higher deposition efficiency (Oberdörster et al., 1994). The absorption of UFP occurs primarily through the lungs but may also take place anywhere on the body surface. Due to their small size, UFPs are capable of penetrating the lung's epithelium and may, therefore, enter the bloodstream. From there, the particles can reach the liver, bone marrow, brain and heart leading to a systemic inflammation.

Particles may deposit in the lung by one of four methods, namely: (a) interception, (b) impaction, (c) sedimentation and (d) diffusion (CCOHS, 1999).

(a) Interception occurs when a particle moves so close to the airway surface that its edge touches the surface. This method of deposition is commonly seen with fibres, such as, asbestos.

(b) Particles tend to travel along their original course. In case of a bend within the airway, particles impact with the airway surface in their path rather than turning with the flow of air. This deposition, also known as impaction, is dependent on the air velocity and particle mass.

(c) Sedimentation takes place when gravitational forces and air resistance overcome the tendency of the particle to remain suspended in air. As a result, the particles deposit on a lung surface. Sedimentation is common in the bronchi and bronchioles.

(d) Diffusion, or random movement, occurs with particles having an aerodynamic diameter smaller than 0.5µm. When particles are in random motion, they deposit on the lung walls mainly by chance.

1.3. Particulate Matter and Health Effects in Epidemiological Studies

Numerous studies have shown that high exposure to fine and ultrafine particles may have adverse long- and short- term effects on human health (Brunekreef and Holgate, 2002). The following section presents adverse health effects related to PM and UFP exposure according to the system affected and sorted by long- and short-term effects.

Respiratory System

An increased respiratory morbidity and mortality was found in sensitive populations (Wichmann et al., 2000). Furthermore, there is evidence that particulate matter induces mild focal interstitial fibrosis, chronic bronchitis and exacerbation of asthma

(Baggs et al., 1997; Brown et al., 2002; Holgate et al., 2003). Holgate et al. (2003) also found changes in lung function and histology along with inflammation and respiratory tract infection (Osunsanya et al., 2001; Brown et al., 2002). These lung conditions account for an increase in hospital admissions and a resultant absence from work and school.

Cardiovascular System

It has been shown that cardiovascular morbidity and mortality is increased when subjects are exposed to high concentrations of particulate matter (Wichmann et al., 2000). A higher incidence of ischaemic heart disease and myocardial infarction has also been found, and Pekannen et al., (2002) also found an increase in other heart conditions as well as cardiovascular damage.

Circulatory System

UFPs have been shown to penetrate the interstitial space very deeply and rapidly in and are able to enter the bloodstream. Furthermore, a change in blood parameters also has also been shown (Henriksson and Tjalve, 2000).

Nervous System

Besides causing alterations in the autonomic nervous system, such as, changes in arterial blood pressure which is a cardiovascular risk factor, particle accumulation was observed in the olfactory bulb (Dorman et al., 2004).

Apart from the specific conditions mentioned above, several other symptoms have also been identified on exposure to particulate matter. These include cough, fatigue, muscle aches, neck discomfort and various allergies (Frampton et al., 1992).

During two large studies in Europe and the USA (APHEA Air Pollution and Health: a European Approach; NMMAPS National Mortality and Air Pollution Studies) investigating the short term effects caused by particulate matter, it was shown that asthma, chronic obstructive pulmonary disease and daily mortality increased with an increase in PM_{10} levels (Touloumi et al., 1997; Atkinson et al., 2001; Roberts and Martin, 2006; Katsouyanni et al., 2009). There is also further evidence showing that cardiac deaths, and various cardiac and respiratory complications can arise leading to an increase in hospital admissions (Peters et al., 1997; Peters et al., 1999a). An increased incidence of lower respiratory symptoms including cough as well as exacerbations of asthma and a decline in lung function was also noted (Pope, 2000). Epidemiological evidence furthermore suggests that PM may have an influence on heart rate variability resulting in a higher risk of cardiac arrhythmias (Devlin et al., 2003; Gong et al., 2003; Gong et al., 2008).

With regard to long-term effects, a higher mortality rate was found in areas with higher particulate pollution levels. Animal studies have also supported epidemiological evidence that different types of PM, in particular diesel exhaust particles, induce the development of cancer in human beings (McClellan, 1987; Pope et al., 2002; Schins, 2002). An increase in respiratory and cardiovascular mortality has also been found in adult subjects (Dockery et al., 1993; Pope et al., 1995; Hoek et al., 2002; Pope et al., 2002). Prolonged exposure of infants to these particles has resulted in respiratory arrest and sudden infant death syndrome (Woodruff et al., 1997; Woodruff et al., 2006). Studies have additionally shown an increase in chronic cough, bronchitis and other chest ailments in subjects living in areas with higher levels of PM pollution (Abbey et al., 1995; Dockery et al., 1996; Peters et al., 1999b). A general decline in lung function has also been associated with exposure to high levels of particulate matter (Pope, 2000). In addition to this, it has been suggested that long-term exposure to PM pollution is also associated with higher levels of fibrinogen in the blood as well as elevated platelet and white blood cell counts (Schwartz, 2001). Künzli et al., (2005) and Hoffmann et al., (2007) have also shown a link between PM exposure and atherosclerosis. All these, in turn present a higher cardiovascular risk.

To date it has still not been identified which specific physical or chemical characteristics of PM are responsible for the adverse health effects produced. Although toxicology studies carried out on animals suggest that ultrafine particles are more toxic than a similar dose of larger particles (Oberdörster, 2001), it is unclear whether these health problems are caused by particle size alone or by a combination of effects caused by their chemical and biological components, by specific organic compounds, or by certain gases (Schlesinger et al., 2006). However, due to these adverse health effects, it becomes obvious that assessing PM and UFP exposure is the keystone to understanding dose and effect relationships as well as to protecting human life from potential risks.

1.4. Central Site Exposure Assessment in Epidemiological Studies

The basic concepts used in exposure assessment were developed in the early 1980s by Duan (1982) and Ott (1982). Their introduction of the term 'human exposure' emphasises that the human being is the most important receptor of pollutants in the environment. Ott (1982) elaborated a system of definitions for the term 'exposure' and defined exposure as "an event that occurs when a person comes in contact with the pollutant". This is a definition of an instantaneous contact between a person (*i*), or a group of persons, and a pollutant with concentration (*c*), at a particular time (*t*).

Epidemiological studies in the field of short-term exposure focus on daily or other acute variations in pollution level and linked health endpoints. Short-term exposure studies investigating particle exposure and subsequent health effects are based on short-term temporal variations using concentrations at a central site or other stationary or mobile sampling devices.

Chronic or long-term exposure studies focus on health endpoints across communities at different pollution levels over time periods of one year and longer. Early long-term epidemiological studies investigating the association between particle exposure and health effects are based on a central monitoring site located in an urban background. A central issue in exposure assessment is to study the spatial variation, namely, how well particle concentrations in a wider urban area correlate with values obtained when measuring concentrations at a single centrally located site.

1.4.1. Central Site Measurement

Early epidemiological studies on air pollution obtained exposure data from ambient monitoring networks (Dockery and Pope, 1994; Schwartz, 1994; Pope et al., 1995; Pope et al., 2009). In these studies, people living in defined areas, such as, in a particular city, were assigned to the same pollution concentration. These studies typically used air pollution measurements from stationary air monitoring sites within a community as a surrogate for personal exposure levels for the population. Ambient monitoring networks have been established all over Europe and the United States of America by national institutions or local councils and are equipped with online monitors providing continuous data.

1.4.2. Measurement of Personal Exposure

In order to develop personal exposure models, it is important to measure personal exposure during different exposure situations. The following section will, subsequently, introduce different sampling methods.

Among personal sampling devices, passive samplers are the most widespread and easily used devices employed in personal sampling. They exist as small tubes or badges and rely on the principle of passive diffusion of a gas (Palmes et al., 1976). The use of these samplers is very straightforward as they are lightweight, do not require a source of electricity, and can be easily attached to outer clothing. Furthermore, passive samplers may also be used to take stationary measurements in outdoor and indoor settings.

The most accurate way of determining personal exposures is by means of monitoring devices such as active integrating samplers which incorporate filters through which air is pumped at fixed flow rates. Instruments, such as, the Langan CO Personal Exposure Measurer (Langan Products, Inc., San Francisco, CA, USA) and the TSI Model 8510 piezobalance (TSI, Inc., Minneapolis, MN, USA) may also be used in order to collect real-time data (Klepeis et al., 2007). Personal monitors for particles exist with different cut-off points and size fractions, such as, UFP, PM_1, $PM_{2.5}$, PM_4, PM_{10}, or multistage samplers which collect particles using various filters. These particles are then analysed by means of gravimetry.

Many studies focussing on personal UFP sampling made use of a microenvironmental based sampling technique for a variety of reasons utilising a real-time condensation counter, namely the handheld CPC 3007 by TSI, St. Paul, MN, USA. This was done for a variety of reasons, namely mobility and user-friendliness. (Diapouli et al., 2007).

1.5. Different Exposure Assessment Methods - Advantages and Disadvantages

Modelling simulates exposures using:
(i) empirical distributions of exposure in specific microenvironments,
(ii) output from microenvironmental models, and
(iii) human activity pattern data.

Modelling has the main advantage of offering rapid and inexpensive exposure estimates over a wide range of exposure scenarios. In contrast to modelling exposure, the sampling of exposure involves the deployment of a large number of exposure monitors. In the sampling approach, different exposure scenarios must be investigated by collecting additional data (Klepeis, 1999). It is known, however, that particle concentrations at a central site can correlate rather poorly with personal exposure to particles on a given day as personal exposure is influenced by several factors besides ambient-air concentration. Such factors include indoor penetration, indoor sources, and activity during leisure time (Pekkanen and Kulmala, 2004).
A main disadvantage of modelling is the need for systematic validation. This means that the results of exposure assessment have to be compared to an independent set of directly-measured exposure data. The data-intensive nature of this approach has made validation difficult as individual human activity patterns need to be taken into account (Ott et al., 1988). The availability of new activity patterns and other exposure-related databases is encouraging (Sexton et al., 1995; Jenkins et al., 1996; Wilson et al., 1996).

Exposure assessment for UFPs is still in the initial stages compared to exposure assessment for fine particles $PM_{2.5}$ and PM_{10} (Pekkanen and Kulmala, 2004; Sioutas et al., 2005). When compared to other particulate matter, UFPs have shorter atmospheric lifetimes in the order of hours, and this can be even shorter in the vicinity of local particle sources with higher UFP concentrations. They are also transported over shorter distances. With growing distance from the particle source, both atmospheric dilution and coagulation play an important role in the rapid decrease in particle number. Due to their different physical properties as described above, UFPs are expected to have larger spatial and temporal variability than fine particles. This is, in fact, seen as a less even distribution within a particular area when compared with other particles (Pekkanen and Kulmala, 2004). Compared to exposure assessment of mass concentration of $PM_{2.5}$ or PM_{10}, exposure assessment of UFP based on values measured at one single monitoring site is, therefore, more error-prone (Monn, 2001; Pekkanen and Kulmala, 2004). Moreover, not many studies on temporal and spatial correlation of UFP across an urban area are available to date (Buzorius et al., 1999; Aalto et al., 2005; Hussein et al., 2005; Tuch et al., 2006; Puustinen et al., 2007).

In summary, none of the methods developed so far have been capable of accurately assessing long-term personal exposure to UFP. Several shortcomings include the failure to evaluate all UFP-emitting sources within an individual's immediate environment as well as the lack of assessment of the various activities carried out by people within the study population. The first promising approach to assessing all these sources and activities has been through the EXPOLIS study by introducing a microenvironmental based model for short-term exposure to $PM_{2.5}$.

As mentioned previously, UFP may originate from several local sources within the participant's home, such as cooking or cleaning. These sources, especially those within the participant's place of residence, have a major impact on their personal exposure as individuals spend most of their time indoors. Applying models without taking these local UFP sources into account would inevitably lead to erroneous results. It is, therefore, crucial to develop a new microenvironmental-based model for the assessment of personal exposure to UFP incorporating these local sources.

2. Goals of this Study

Exposure-related modelling is a tool allowing assessment of exposure and dose in situations where all the relevant measurement data is unavailable. In this sense, exposure-related models are tools for extrapolating available information to different situations without requiring complete measurement in each new situation. This is clearly an important use due to the very high costs entailed in obtaining measurement data (Furtaw, 2001).

Until now, the development of models focussed on ambient sources rather than on local or even personal UFP-emitting sources. This study was, therefore, designed to focus on the development of an exposure model which incorporates personal information on a level of detail beyond that of other studies. The purpose of this research is to develop a personal exposure model that can be used to estimate personal exposure to ultrafine particles by means of utilising simple input data, such as, time spent in various microenvironments and exposure to specific sources. The specific aim of this study is to develop and validate a personal exposure model which can be used to estimate UFP exposure levels in a large cohort. This specifically includes the following steps:

1. Literature review and identification of specific exposure situations for high and low UFP exposure including identification of factors influencing personal exposure, such as, specific behaviour and habits or local sources of emission.

2. Definition of specific microenvironments which are associated with UFP exposure, namely in-traffic, residential-indoor, residential-outdoor and at-work.

3. UFP exposure assessment

   3.1. Personal exposure measurements for 22 participants by means of a handheld CPC (TSI 3007) personal sampler
   3.2. Development and implementation of a personal diary
   3.3. Allocation of sampled data to diary statements
   3.4. Identification of specific periods of exposure and allocation of sampled data
   3.5. Development of a questionnaire for collecting information on personal habits, home specifications and the use of transportation, such as, car, bicycle and public transport.
   3.6. Identification of main contributors to UFP concentration and time spent performing each activity
   3.7. Comparison of diary and questionnaire data

4. Model development

   4.1. Development of literature database for UFP sources and exposures
   4.2. Development of a model:
        a) based on 48-hour diary data (short-term)
        b) based on questionnaire data (long-term)
   4.3. Calculation of mean exposure for each microenvironment
   4.4. Calculation of mean total 24 hour exposure for each participant

3. Materials and Methods

This section is subdivided into three subsections, namely (i) preparation steps, definition, and development of diary and questionnaire (ii) UFP sampling and (iii) data analysis and modelling.

I. Preparation Steps, Definitions and Development of a Diary and Questionnaire

3.1. Literature Search

A systematic literature search of electronic databases, namely PubMed and Google Scholar, was performed. The search was supplemented by references from relevant articles. Keywords for searches were: personal exposure, UFP modelling, microenvironmental modelling and a combination of UFP sources mentioned in the questionnaire. As the scope of this thesis was to design a pilot study targeting a specific area, no comparison was made with UFP readings obtained from other countries or localities. Should the pilot study be adapted for use on a larger scale, then it would be of value to investigate the comparability of the readings obtained in different countries.

3.2. Study Population

Participants were recruited after a personal interview. All participants were personally known to the candidate. Volunteers had to be able to understand German in order to complete the questionnaire and diary and to be able to understand the instructions. Furthermore, participants had to be over 18 years of age and non-smokers. People unable to carry TSI handheld CPC 3007 due to medical reasons or restrictions at the workplace were excluded from the study.

3.3. Definition of Microenvironments

In order to develop an exposure model, microenvironments were defined. Definitions for all microenvironments are summarized in Table 3.1.

Tab. 3.1 Definition of Microenvironments visited by the participants within the 48-hour sampling period

Microenvironment	Abbreviation	Definition
Residential-outdoor	RO	Outside the participant’s home
Residential-indoor	RI	Within the participant’s home
In-transit	IT	Method of transportation
At-work	AW	At the work place
Other places	OP	Locations visited during the 48-hour sampling period, which have not been covered by the questionnaire

The following five microenvironments were defined: residential-indoor, residential-outdoor, in-transit, at-work and other places. The latter includes both indoor and outdoor locations. The residential-indoor (RI) ME comprised all activities and sources recorded within the participant’s home or flat, for example, cooking, cleaning and candle burning. In order to take diurnal variation of ambient particles and the variation of activity of indoor sources into account, the RI ME was subdivided into:
(a) day- and
(b) night-time exposure during sleep.
Night-time was defined as the time when participants went to sleep. The beginning and end of this sleeping period was specified by the participants in their diary (N=19). In case of missing information, sleeping time was assumed to commence at 23:00 and end at 08:00 (N=2). In case of missing information and active UFP-emitting sources after 23:00, the next possible starting point without active sources was chosen (N=1).

The residential-outdoor (RO) ME comprises all data collected at the participant’s home address as well as outside the home within a 5 km range, such as, spending time in the garden or walking in the neighbourhood. The in-transit (IT) ME includes all measuring periods that are associated with transportation, including driving a car, riding a bicycle, walking, or travelling by means of public transportation. Different ventilation modes while travelling by car, such as, windows opened or closed, ventilation system on or off, were recorded. All participants, however, claimed to travel with closed windows and activated ventilation system. The at-work (AW) microenvironment focussed on the time spent at the work place. Working activities and UFP sources at the work place, such as, laser printers (He et al., 2007) and fuel combustion, were recorded. Any activity or location which did not fall into one of the above-mentioned MEs was included into the category 'other places' (OP). Some examples would be, time spent in pubs or meeting friends at locations other than the home address.

Within each microenvironment, specific periods of personal activity likely to modify the exposure were recorded, namely, cooking, cleaning, candle burning, the use of heaters as well as different methods of transportation. These periods were defined by their predominant source of exposure, such as, cooking or cleaning. A specific period, such as, cooking, is a period of time that is measured without interruption during a specific UFP exposure situation.

3.4. Questionnaire for the Collection of Long-Term Personal Information

The questionnaire was designed to collect information regarding long-term personal activity and behaviour, such as, means of transportation, room ventilation, exposure to environmental tobacco smoke, cooking as well as candle burning. Questions asked and answer options translated from the original German questionnaire are shown in Table 3.2. Questions attempted to collect data reflecting all areas of daily life in order to provide sufficient information for long-term modelling. Time-based questionnaire statements were transferred into electronically processable time units as follows:
(a) occasionally: 15 minutes,
(b) less than 30 minutes: 30 minutes,
(c) 30 minutes to 2 hours: 75 minutes and
(d) more than 2 hours: 120 minutes.

Tab. 3.2 English translation of the questionnaire for the assessment of average personal behaviour and activity information

Question	Answer options
estimation of daily distance walked	(a) less than 1 km, (b) 1-2 km (c) more than 2 km, (d) denied (e) unknown
ETS[1] exposure at workplace	(a) yes, (b) no, (c) not applicable, (d) unknown
ETS[1] exposure at home	(a) yes, (b) no, (c) not applicable, (d) unknown
ETS[1] exposure at other places (besides home and workplace)	(a) yes, (b) no, (c) not applicable, (d) unknown
number of smokers at home	____, 88[3] = not applicable, 99[3] = unknown
linear distance of home from busy road	(a) less than 10m, (b) 10-50m (c) more than 50m
home situated in a "street canyon"[2]	(a) yes, (b) no
if above question = "no", which of the following best describes your street/ house	(a) detached houses on both sides of the road (b) at least one side with detached or semi-detached houses (c) house or flat at a crossing (d) other
level of main living room	(a) ground-level, (b) first floor, (c) second floor (d) higher
time spent at home	work days in hours summer:___/winter:___ Sundays in hours summer:___/winter:___
windows open or closed while in living or bedroom	living-room: summer: open/closed; winter: open/closed bedroom: summer: open/closed; winter: open/closed
ventilation of room being used	summer: ___times a day or permanent winter: ___times a day or permanent
daily time spent in rooms with ETS (at work and private time)	summer: hh:mm ____ winter: hh:mm ____
usage of heaters	yes/no
if yes, which of the following applies:	tick any option that applies; rooms: kitchen, living room, bedroom, other type of heater: wood, coal, oil, gas, electricity
location of room	choose any one answer that applies: rooms: living-room, bedroom location: motorway, main road, side road, courtyard (outdoor), courtyard (indoor), park
type of windows	choose any one answer that applies: windows: noise reducing, double-glazed, standard, other rooms: living-room, bedroom
average time spent cooking or in same room	hh:mm: ____
type of cooker	(a) gas, (b) electric, (c) other
use of cooking hood	yes/no
average time spent cleaning per day or in room being cleaned	(a) 0, (b) up to 30 min, (c) 30 min to 2h (d) more than 2h
average time spent burning candles per day	(a) 0, (b) up to 30 min, (c) 30 min to 2h (d) more than 2h
mode of transportation and time spent commuting	choose any one answer that applies: car & lorry, motorcycle, bus & tram, train, on foot, bicycle, subway time spent: not applicable or occasional, less than 30 min per day, 30 min to 2h per day, more than 2h per day
ventilation while driving car	choose any one answer that applies: summer, winter ventilation: windows open, ventilation & air-conditioning, no ventilation
distance of workplace from busy road	(a) less than 10m, (b) 10-50m, (c) more than 50m
activities at work	yes/no (a) soldering, (b) moulding (c) brazing, (d) metal-processing (e) handling of bulk materials (f) working in a garage

[1] environmental tobacco smoke, [2] a street with terraced houses on both sides, [3] for internal use only

3.5. Diary for the Collection of Short-Term Personal Information

The diary was designed to gain short-term information on personal behaviour and activities during UFP measurement periods. The diary, an extract of which is shown in Fig. 3.1, was designed to collect information at 15-minute intervals during the 48-hour sampling period. Information was used for the development of a personal short-term exposure model. Information obtained also served as a means of allocating measured UFP exposure to defined sources and activities. Participants could check which of the activities or sources listed in Tab. 3.3 were applicable. Furthermore, each participant was able to provide additional information if none of the listed activities was appropriate or the sampler's alcohol cartridge or battery pack was replaced. Fig. 3.1 is an extract from the German version of the diary, the full version of which can be found in Appendix II.

Tab. 3.3 English translation of the short-term diary (multiple choice)

Time	In-traffic	At-home	At-work
15-min intervals	(a) walking	(a) cooking	(a) bulk materials
	(b) motorcycle	(b) cleaning	(b) metal-processing
	(c) car/lorry	(c) candle burning	(c) ETS
	(d) bus/tram	(d) ventilating	
	(e) subway	(e) ETS	
	(f) train		

Datum:			AUFENTHALTSORT													
Name:			Unterwegs							zu Hause						
Stunde	Minute	Kurze Beschreibung der Aktivität und des Aufenthaltsortes falls abweichend	Laufen	Motorrad	Auto / LKW	Bus / Staßenbahn	U-Bahn	Zug	Fahrrad	Aufenthalt in verrauchten Räumen	Kochen	Putzen	Kerzen brennen	Lüften
0	0		O	O	O	O	O	O	O	O	O	O	O	O
	15		O	O	O	O	O	O	O	O	O	O	O	O
	30		O	O	O	O	O	O	O	O	O	O	O	O
	45		O	O	O	O	O	O	O	O	O	O	O	O
1	0		O	O	O	O	O	O	O	O	O	O	O	O
	15		O	O	O	O	O	O	O	O	O	O	O	O
	30		O	O	O	O	O	O	O	O	O	O	O	O
	45		O	O	O	O	O	O	O	O	O	O	O	O

Fig. 3.1 Extract from the original German diary-layout as handed out to participants

3.6. Processing of Questionnaire and Diary Data

Completed questionnaires were stored in a MS Access© database. The personal 48-hour diaries were transferred into MS Excel© sheets.

II. UFP Sampling

3.7. Sampling Period and Weather Conditions

Sampling took place from April to July 2008. Temperature values during sampling time ranged from 8 to 20°C (mean 15°C, SD=3°C, median 16°C). Relative humidity mean was recorded at 70%, (SD=11.5%, range 40.5–96%, median 70%). Atmospheric pressure ranged from 1,001 to 1,027hPa with a mean of 1,014hPa (SD=5, median 1,014). All environmental data was obtained from the weather station closest to each participant's place of residence.

3.8. UFP Sampling

The following section will briefly introduce the handheld condensation particle counter, TSI CPC 3007 and highlight the equipment and procedures used.

3.8.1. Introduction of UFP-Sampler TSI CPC 3007

UFPs were monitored by a handheld CPC shown in Figures 3.3 and 3.4. The CPC has been used extensively for indoor and outdoor monitoring in various studies (Matson, 2005; Monkkonen et al., 2005). A built-in pump draws aerosol samples through the instrument with a flow rate of 700cm³/min. Upon entering the unit, the sampling flow becomes saturated with alcohol vapour. Alcohol condenses onto particles larger than 10nm and the resulting droplets are counted optically. The concentration accuracy is ±20% up to 100,000 #/cm³. The lower detection limit by particle diameter (dp_{50}) of the CPC is about 15nm, while the upper limit, mainly due to the CPC design, is about 1µm. The detection limit by particle count is equivalent to 100,000 #/cm³ (TSI, 2007; Asbach et al., 2009). The instrument switches to scattered light mode when UFP concentration is above 100,000 #/cm. The TSI 3007 CPC suffers only weak losses in counting efficiency at values between 100,000–400,000

#/cm³ (Hämeri et al., 2002; Asbach et al., 2009; Cattaneo et al., 2009), therefore, values above 100,000 #/cm³ were included in the analysis (periods N=30).

Particle number concentrations are dominated by UFP and only a very small fraction of larger particles (less than 10%) contributes to the total particle number concentration in urban environments (Sioutas et al., 2005). Therefore, it may be assumed that particles counted by the TSI CPC (0.01-1µm) correspond to the ultrafine size range. A schematic flow together with technical specifications can be found below (Fig. 3.2 and Tab. 3.4). Images of the particle counter are presented in Fig. 3.3 and 3.4.

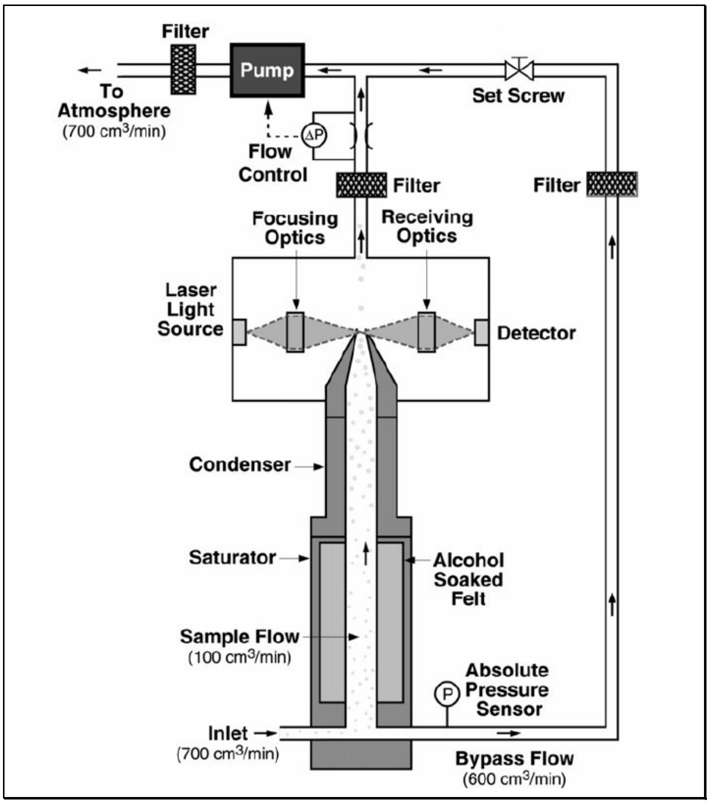

Fig. 3.2 Flow-schematics of TSI handheld CPC 3007 (TSI, 2007)

Tab. 3.4 TSI Handheld CPC 3007 specifications

Particle Size Range	0.01-1μm
Concentration Range	0-100,000 #/cm^3
Minimum Displayable Concentration Value	1#/cm^3
Concentration Accuracy	±20%
False Background Counts	<0.01 #/cm^3
Response Time	<9s for 95% response
Particle Type	Airborne solids and non-volatile liquids
Temperature Range (operational)	10-35°C
Flow Rate:	
Detected Aerosol	100cm^3/min
Inlet	700cm^3/min
Absolute Pressure Sensor	0-1,400hPa
Power Requirement:	
Battery Type	6 AA alkaline or rechargeable
Battery Life	5 hours (alkaline batteries at 21°C)
Alcohol Requirement	100% reagent-grade isopropyl alcohol

Fig. 3.3 The TSI handheld CPC 3007 – Side view

Fig. 3.4 The TSI handheld CPC 3007 – Rear view

3.8.2. Equipment and Software

A list of the equipment and software used for UFP-sampling can be found in Tab. 3.5 and 3.6 below.

Tab. 3.5 UFP sampling equipment

Equipment	Brand/Manufacturer
HEPA filter	TSI, USA
TSI CPC 3007	TSI, USA
6 AA alkaline batteries	various brands
AC adapter	TSI, USA
Isopropanol (min. 95%)	Carl Roth, Karlsruhe, Germany

Tab. 3.6 Software used for data processing purposes

Software	Version	Manufacturer
Office Word©	2003/2007	Microsoft©, USA
Office Excel©	2003/2007	Microsoft©, USA
Office Access©	2003/2007	Microsoft©, USA
Aerosol Instrument Manager©	5.5.2.0	TSI, USA
Photo-Paint X3	13.0.0.576	Corel Corp., USA
MultiTransAHO	4.3	H. Rauschenberger, AHO, Germany
Google Earth©	4.3	Google Inc., CA, USA
SAS	9.1	SAS Institute Inc., Cary, NC, USA.

3.8.3. Preparation and Use of the TSI CPC 3007

Before and after each 48-hour sampling period proper functioning of the TSI handheld CPC 3007 was ensured according to the manufacturer's recommendations. This included checks on the pump, batteries, settings and data-storage. After the initial self-test, lasting 600 seconds, a zero check was performed by using a high efficiency particulate air (HEPA) filter. The device was considered to be functioning normally if a zero count was obtained.

The participants were instructed to place the sampler at a maximum distance of 1m within their breathing space, with the exception of sleeping hours. The influence of distance between sampler and participant was not investigated in this pilot study, therefore, results have not been adjusted in any way. Participants who were disturbed by the high noise level of the in-built pump during the night, were allowed to place the sampler in another room. Doors between the bedroom and this other room could be left open or closed depending on the participant's tolerance to the noise level. The participants were instructed to keep all windows closed during the night. The bedroom or alternative room were not to contain any active UFP-emitting sources, such as, cooking or candle burning. Periods with opened windows during the night were excluded from this analysis (3 nights in 2 subjects). Participants were instructed not to place the TSI sampler on the floor at any time in order to prevent suction of dust and particles from the floor.

3.8.4. Data Management

Data was collected according to the manufacturer's guidelines using the Aerosol Instrument Manager©. The data was then exported into a text file and finally converted into an Excel© sheet. Data was then sorted according to date and time stamp as indicated in Table 3.7 below. A floating mean was used to calculate 15 minute intervals in order to make acquired diary data comparable to the model's calculations.

Tab. 3.7 Allocation of time-stamps as obtained by the TSI sampler

Time stamp according to sampling device [hh:mm:ss]	Corresponding time stamp in the diary [hh:mm:ss]
14:15:01 or later	14:15:00
14:29:24 or closer to 14:30:00	14:30:00
14:46:26	14:45:00
14:55:08	15:00:00

III. Data Analysis and Modelling

3.9. Data Analysis

All date collected for each participant was converted into MS Excel sheets and calculations, box-plots (whiskers: min and max, bottom and top of the box: 1st and 3rd quartile, band: median) and tables were generated in MS Excel 2003©. The raw data and corresponding diary statements were also visually inspected in order to identify implausible recordings. A sudden rise or drop of more than 90% of the recorded concentration for at least 5 minutes without an accompanying specification in the diary was excluded from the analysis of MEs and specific exposure situations (11.7 hours were excluded from a total sampling time of 936 hours). Data was grouped into microenvironments as stated above (RI, RO, OP, AW and IT) and 5-minute mean exposures were calculated based on raw data taken every 30 seconds. Data within MEs was subdivided into specific exposure situations of defined activities/sources as stated by the participant. Time periods which did not allow allocation to a specific ME due to insufficient data were excluded from the analysis. This included failure of the measurement device or incomplete diary information.

From the 5-minute averaged exposure data, the mean, SD, median and 1st and 3rd quartile were calculated for each ME and specific exposure situations of cooking, cleaning and travelling by car. In order to estimate the variability of measurements within single periods of active sources, the coefficient of variation (*CV*) for participant (*i*) in period (*j*) (CV_{ij}) according to the 5-minute readings was calculated using the following equation:

(eq. 1) $$CV = \frac{\text{standard deviation}}{\text{5 min mean exposure}}$$

For the assessment of variation of exposure within a person across all specific periods of a kind, such as, all periods of cooking in participant *i*, the CV_i from all 5-minute readings during all specific periods of one kind was calculated. CV_{tot} is based on all periods of a kind across all participants and reflects the total variation of exposure for a specific exposure situation within this study population. The CV's were calculated in order to investigate how consistent and comparable same type periods are within and across participants.

In order to estimate the additional exposure due to a specific source, such as, cooking or cleaning, baseline values were derived from preceding time periods when no specified active source was recorded (reference periods). The difference between the reference period and the index period with active emitting source was calculated.

In case of missing reference periods, the index period with active source was not taken into account for this analysis. The assessment of the increase in exposure in the in-transit ME was performed analogous to the residential-indoor microenvironment. Reference periods were obtained from the residential-indoor ME as no reference periods within the in-transit microenvironment could be obtained. Periods within the RI ME in the absence of UFP-emitting sources are the closest reference periods that could be obtained during this pilot study.

3.9.1. Identification of Main Contributors

The aim of this analysis was to assess the contribution of the three most frequent specific exposure situations, namely, periods of cooking, cleaning and travelling by car on the cumulative daily UFP exposure of the participants. Since the diary only had a 15-minute time resolution, the 15-minute mean exposure periods were used rather than the 5-minute mean exposure periods. Cumulative UFP exposure (*E*) for each participant (*i*) during the complete sampling period of approximately 48 hours was calculated using equation 2:

(eq. 2) $$E_i = \sum_{m=1}^{n_i} x_m \left[\frac{\#}{\mathrm{cm}^3}\right]$$

where x_m are the 15-minute mean concentrations of the m^{th} interval, and $m=1,\ldots,n_i$. Cumulative UFP exposure ($E_{i,j}$) for the exposure periods (*j*) (cooking, cleaning or travelling by car) was calculated for each participant (*i*) according to equation 3:

(eq. 3) $$E_{i,j} = \sum_{k=1}^{n_{ij}} x_k \left[\frac{\#}{\mathrm{cm}^3}\right]$$

where x_k are the 15-minute concentrations during the k^{th} period of specific UFP exposure ($k=1,\ldots,n_{ij}$). For each period (*j*) the cumulative concentrations ($E_{i,j}$) were averaged over all participants who were exposed at least once to the specific UFP source within the 48 hours. The percentage of UFP exposure during specific exposure situations, such as, cleaning, cooking and travelling by car, of the total cumulative exposure was then calculated using equation 4:

(eq. 4) $$\frac{E_{i,j}}{E_i} \cdot 100 = CB_{ij}$$

where CB_{ij} is the contribution of the specific exposure situation (cleaning, cooking and travelling by car) to the total exposure in participant (i). The percentages (CB_{ij}) for all participants who were exposed to the specific UFP source for each (j) were then averaged. Overall sampling time for each participant (i) (T_i) and sampling time during the exposure periods of cooking, cleaning and travelling by car (T_{ij}) were calculated for each participant in an analogous fashion. The percentage of time spent in each exposure situation was calculated according to the equations for the UFP exposure for each participant. The average over all participants who were exposed to the specific UFP exposure was then calculated.

3.9.2. Comparison of Questionnaire and Diary Statements

Information gained from the questionnaire is regarded as reference data. Participants were requested to provide information on the average time spent in specific microenvironments. Information from the diary is regarded as control data for a specific day (sampling time) that may not reflect the participant's usual behaviour. Information from the questionnaire and diary were compared in order to evaluate whether data gained during sampling reflects the participant's average behaviour. Deviations between both diary and questionnaire information were calculated for each participant and for the entire study population. Furthermore, differences between both data sets were highlighted and presented as charts.

3.9.3. Comparison of Measured and Literature Exposure Values

In order to verify measured data, sampled data was compared to literature findings. Verification was performed by determining whether measured data was within the range of literature values.

3.10. Development of a Personal UFP Exposure Model

It may be assumed that personal exposure *(PE)* is the sum of all UFP-emitting sources and activities within all microenvironments visited during a 24-hour period. Therefore, the exposure during each activity and exposure to a specific source must be calculated. The calculation is based on a given time of exposure and a given UFP concentration obtained from literature. A mathematical modelling approach can be found in the results section.

4. Results

4.1. Definition of Microenvironments Including Influencing Factors

This study was based on a microenvironmental approach so as to develop a model for ultrafine particle exposure. The model was developed from data obtained from a questionnaire defining the participant’s home surroundings, such as, proximity to streets and their personal behaviour and habits, including passive smoking, candle burning and cooking.

Figure 4.1 summarises all sources and influencing factors covered by the questionnaire for the residential-indoor microenvironment. Many sources may be influenced by modifying factors, such as, cooking by the use of an exhaust hood. Infiltrating outdoor UFP concentration is influenced by the surrounding environment as well as by other factors, such as, ventilation. The RI microenvironment may be regarded as the most important microenvironment as people tend to spend most of their time indoors. A high exposure in the RI microenvironment automatically results in an overall higher exposure due to the increased amount of time spent within this microenvironment.

Residential-indoor Microenvironment

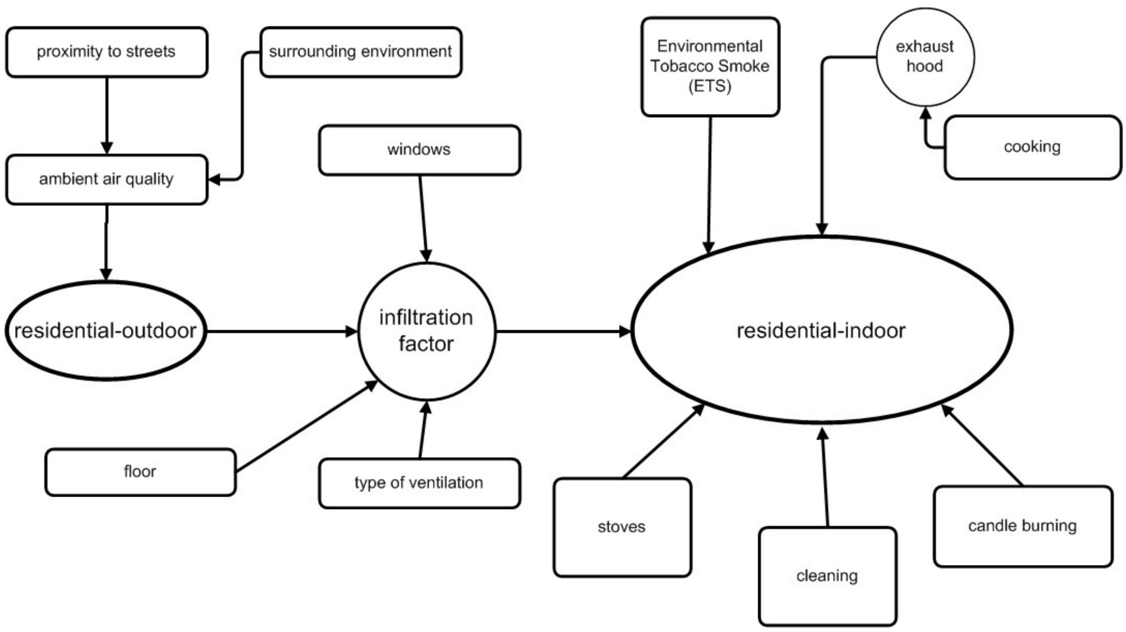

Fig. 4.1 Schematic description of the residential-indoor microenvironment, including important indoor and outdoor sources

In-traffic Microenvironment

The in-traffic microenvironment comprises all means of transportation covered by the questionnaire as well as influencing factors presented by the participant or other passengers within the vehicle. Travelling by car or lorry are the only means of transportation that may be influenced by the participant. Passengers may influence the type of ventilation in various ways, such as, by opening windows, by using the ventilation system, as well as by smoking inside the vehicle. Exposure while travelling by car is highly dependent on the type of ventilation, including: (i) whether windows are open or closed, (ii) the use of the in-built ventilation system and, (iii) the use of cabin air filters.

All other means of transportation cannot be directly influenced by the participant. Such situations include the control of the ventilation system on public transportation as well as the opening of doors at stops or stations. Smoking is prohibited on means of public transportation in Germany and has, therefore, not been taken into account. It should also be mentioned, however, that personal exposure while walking or riding a bicycle may also be dependent on the amount of physical exertion. Riding a bicycle at higher speeds causes much more physical exertion which, in turn, may lead to higher particle deposition in the lungs (Fig. 4.2).

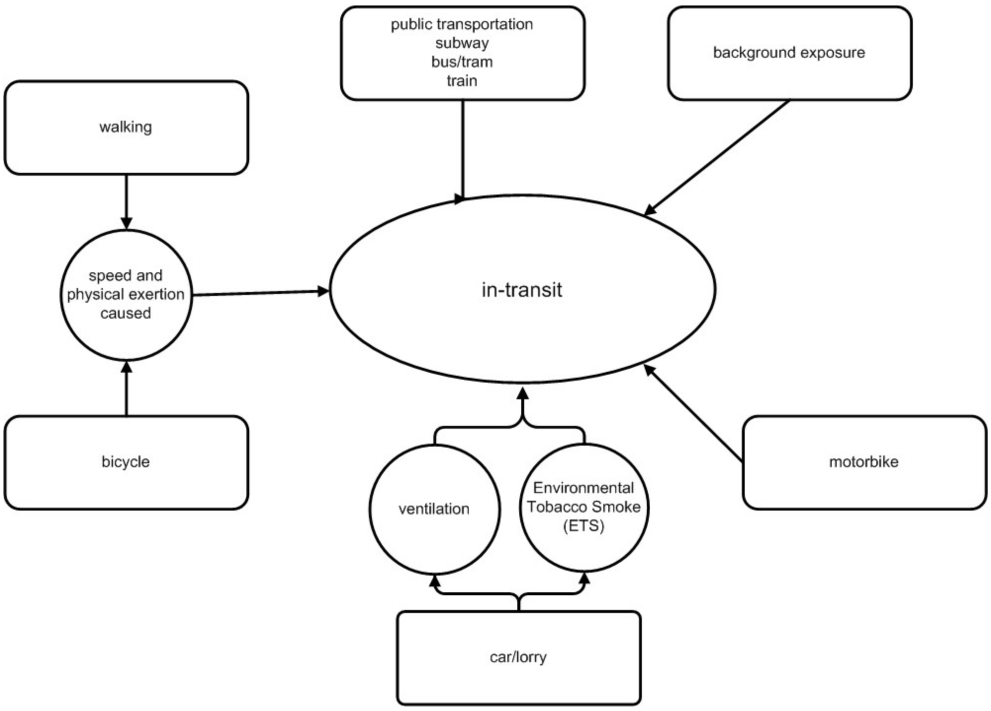

Fig. 4.2 In-traffic microenvironment model including all means of transportation and modifying factors

At-work Microenvironment

The at-work microenvironment is influenced by various sources and activities, such as, soldering and working with bulk materials or any other processes emitting large amounts of dust. The UFP exposure at the work place is also influenced by the ambient UFP concentration and its influencing factors, such as, type of windows and ventilation habits. Office workers may also be highly exposed to UFPs when using laser printers. Specific jobs, namely, coal mining, working in garages and jobs with high exposure to fine aerosols, such as, spray painting, may also contribute to a higher personal UFP concentration (Fig. 4.3).

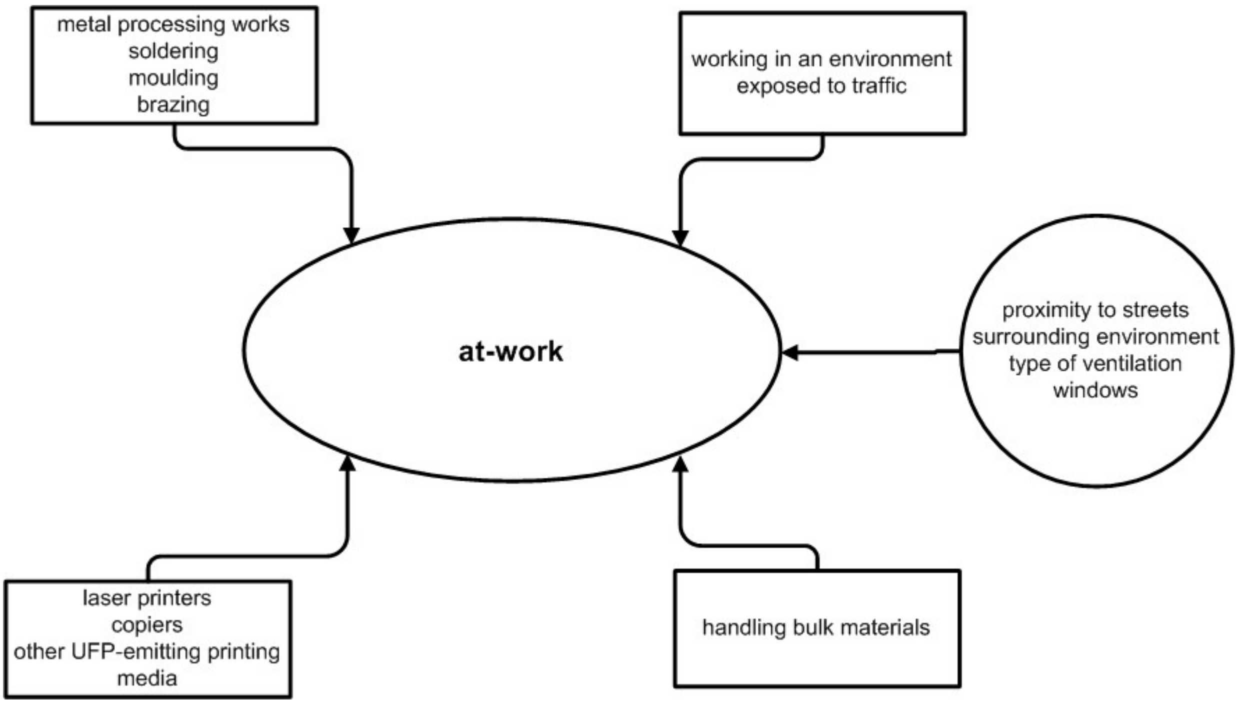

Fig. 4.3 At-work microenvironment model including modifying factors and UFP-emitting sources and activities

The models presented above do not cover all UFP-emitting sources and activities as many other specific UFP sources are still being discovered. However, all sources covered by the questionnaire are presented.

Based on the above-presented ME overviews, the following section is a mathematical approach to developing a personal UFP exposure model.

4.2. Mathematical Modelling Approach

It has been assumed that the daily personal UFP exposure is a time weighted sum of all UFP-emitting sources and activities which the participant is exposed to throughout the day. The total personal exposure for the sampling time (PE) is the sum of each microenvironment: residential-outdoor, residential-indoor, at-work and in-transit per 24 hours (eq. 5):

(eq. 5) $$PE = E_{RI} + E_{RO} + E_{AW} + E_{IT}$$

The exposure for each microenvironment (E_{ME}) is the sum of all UFP-emitting locations and sources ($E_{l/sN}$) within the specified microenvironment (eq. 6).

(eq. 6) $$E_{ME} = E_{l/s1} + E_{l/s2} + E_{l/s3} + \ldots E_{l/sN}$$

For each location or source contributing to UFP exposure ($E_{l/s}$), the corresponding UFP concentration from literature (c_{lit}) is multiplied by the time (t) spent at the location or in the vicinity of the source. In case of more than one literature value for the same source of exposure, the mean of these values was calculated and used. Cumulative time spent in locations and in the presence of specific sources is equal to 24 hours (eq. 7).

(eq. 7) $$E_{l/s} = t \cdot c_{lit}$$

In order to calculate exposure while travelling by car (E_{car}), seasonal changes in ventilation habits like travelling with open windows, using the car's ventilation system or not ventilating at all, ($c_{car\,\mathrm{ventilation}}$, $c_{car\,\mathrm{windows}}$ and $c_{car\,no\,\mathrm{ventilation}}$) must be taken into account. For the purpose of this pilot study, it was assumed that participants travelling by car followed 6 months of winter and 6 months of summer ventilation habits. This means, that statements from the questionnaire concerning the ventilation method while travelling were each applied for 6 months of the year. Literature values were found for measurements while travelling with different ventilation methods (eq. 8).

(eq. 8) $$E_{car} = t \cdot (0.5 \cdot \mathrm{c}_{car\,ventilation\ \mathrm{winter}} + 0.5 \cdot \mathrm{c}_{car\ \mathrm{ventilation\,summer}})$$

Cooking exposure ($E_{cooking}$) is dependent on the kind of energy source used ($c_{lit\,\mathrm{cooking}}$) and an influencing factor extracted from literature - the presence or absence of a fume extractor ($f_{(fume\,extractor)}$). In case of the absence of a fume extractor, the factor is assumed to have a numeric value of 1 (eq. 9).

(eq. 9) $$E_{cooking} = c_{lit\ \mathrm{cooking}} \cdot t \cdot f_{(fume\,\mathrm{extractor})}$$

Exposure to heaters (E_{heater}) is a function of time spent in rooms with active heaters, the type of energy used ($c_{lit\,\mathrm{heater}}$) and the seasonal ventilation habits and frequency of use of the heaters ($t_{indoorssummer}$ and $t_{indoorswinter}$). Exposure data for each kind of heater is extracted from literature. The time spent at home is equal to the time being exposed to heaters. If applicable, time spent in the kitchen ($t_{kitchen}$) as well as sleeping ($t_{sleeping}$) may be deducted. It is assumed that cooking processes are predominant in the kitchen and that no UFP-emitting sources are active during sleep (eq. 10).

(eq. 10) $$E_{heater} = c_{lit\,heater} \cdot \left((t_{indoorssummer} \cdot 0.5 + t_{\mathrm{indoorswinter}} \cdot 0.5) - t_{kitchen} - t_{sleeping}\right)$$

Environmental tobacco smoke exposure (E_{ETS}) is a function of the time spent in locations with ETS exposure ($c_{lit\ \mathrm{ETS}}$) during summer and winter (t_{summer} and t_{winter}) (eq. 11).

(eq. 11) $$E_{ETS} = c_{lit\,\mathrm{ETS}} \cdot (t_{summer} \cdot 0.5 + t_{\mathrm{winter}} \cdot 0.5)$$

The UFP exposure due to ambient UFP concentration ($E_{ambient}$) is a function of the time spent in the RI microenvironment (t_{RI}) multiplied by an infiltration factor (f_{inf}) and the ambient UFP concentration in the participant's home ($c_{ambient}$) (eq. 12).

(eq. 12) $$E_{ambient} = t_{RI} \cdot f_{\mathrm{inf}} \cdot c_{ambient}$$

II. UFP sampling

4.3. Literature Search for specific Sources of UFP Exposure

All results from the literature search will be presented in this section. Table 4.1 and Table 4.3 summarise all literature data for UFP exposure during specific activities also presenting standard statistics, such as, the mean, minimum and maximum values. Modelling was based on this summary approach. Literature findings on infiltration and influencing factors can be found in Table 4.2.

Tab. 4.1 Summary of UFP data from literature including standard statistics

Source	Number of studies	UFP concentration [#/cm³]		
	N	Mean	Min	Max
In-transit				
Bicycle	1	84,005	-	-
Bus	3	29,144	27,825	31,489
Subway	5	20,650	2,667	33,583
Car windows opened	5	17,334	8,216	22,372
Car ventilation system on and windows closed	2	8,919	1,905	15,932
Car ventilation system off and windows closed	1	10,815	10,815	10,815
Walking	6	22,275	14,755	30,334
Train	1	67,115	67,115	67,115
Residential-indoor				
Electric cooking	7	53,441	16,000	150,900
Gas cooking	7	112,714	26,000	146,000
Wood burning	1	17,546	17,546	17,546
Cleaning	9	22,194	550	41,300
ETS	2	119,800	26,600	213,300
Candle burning	4	147,775	69,600	241,500
Electric heater	4	115,941	17,064	218,400
Gas heater	1	79,600	-	-
At-work				
Working in diesel depot	1	142,286	142,286	142,286

Tab. 4.2 Infiltration and ventilation factors extracted from literature

Factor	Numeric value	Reference
Infiltration ($PM_{2.5}$)	0.59-0.71	(Hänninen et al., 2004)
Infiltration (UFP)	0.52	(Hussein et al., 2005)
Infiltration ($PM_{2.5}$)	0.26-0.87	(Wallace and Williams, 2005)
Influencing factor due to the use of an exhaust-hood	0.67	(Still and MacCarty, 2006)

Infiltration factors for $PM_{2.5}$ and UFP obtained from literature vary between 0.26 and 0.71 depending on the house's properties, such as, the type of windows installed, as well as, insulation and ventilation methods.

Tab. 4.3 UFP data extracted from literature, including method, source and standard statistics

Exposure situation	Number of samples N	UFP concentration [#/cm³] Min	Max	Mean	Method	Reference
In-transit						
Bus	-	8,935	66,204	28,029	TSI P-Trak	(Weichenthal et al., 2008)
	-	6,379	83,193	31,489	TSI P-Trak	(Weichenthal et al., 2008)
	36	-	137,350	27,825	TSI P-Trak, TSI CPC 3007	(Hill and Gooch, 2007)
Bicycle	10	41,060	104,952	84,005	TSI P-Trak	(Kaur et al., 2006)
Subway	-	-	-	29,000	TSI P-Trak	(Seaton et al., 2005)
	-	-	-	14,000	TSI P-Trak	(Seaton et al., 2005)
	-	-	-	2,400	TSI P-Trak	(Seaton et al., 2005)
	3	-	10,591	2,667	TSI P-Trak, TSI CPC 3007	(Hill and Gooch, 2007)
	3	-	223,738	33,583	TSI P-Trak, TSI CPC 3007	(Hill and Gooch, 2007)
Car windows open	15	-	185,399	18,747	TSI P-Trak, TSI CPC 3007	(Hill and Gooch, 2007)
	7	-	114,669	8,216	TSI P-Trak, TSI CPC 3007	(Hill and Gooch, 2007)
	16	-	217,838	20,003	TSI P-Trak, TSI CPC 3007	(Hill and Gooch, 2007)
	2	-	120,650	20,131	TSI P-Trak, TSI CPC 3007	(Hill and Gooch, 2007)
	34	-	150,612	22,372	TSI P-Trak, TSI CPC 3007	(Hill and Gooch, 2007)
Walking	-	5,879	89,194	25,161	TSI P-Trak	(Weichenthal et al., 2008)
	-	4,804	40,035	15,778	TSI P-Trak	(Weichenthal et al., 2008)
	46	-	-	30,334	TSI P-Trak	(Briggs et al., 2008)
	6	-	189,982	14,755	TSI P-Trak, TSI CPC 3007	(Hill and Gooch, 2007)
	3	-	227,787	16,665	TSI P-Trak, TSI CPC 3007	(Hill and Gooch, 2007)
Car vents off and windows closed	12	-	60,620	10,815	TSI P-Trak, TSI CPC 3007	(Hill and Gooch, 2007)
Car vents on	13	-	51,888	15,932	TSI P-Trak, TSI CPC 3007	(Hill and Gooch, 2007)
	8	-	7,859	1,905	TSI P-Trak, TSI CPC 3007	(Hill and Gooch, 2007)
Train	17	-	385,152	67,115	TSI P-Trak, TSI CPC 3007	(Hill and Gooch, 2007)

Exposure situation **Residential-indoor**	Number of samples N	Min	Max [#/cm³]	Mean	Method	Reference
Candle burning	-	-	-	69,600	TSI P-Trak	(Afshari et al., 2005)
	-	-	-	241,500	TSI P-Trak	(Afshari et al., 2005)
	-	-	-	66,900	TSI SMPS 3034	(Sun et al., 2007)
	-	-	-	211,000	TSI SMPS 3034	(Sun et al., 2007)
Gas stove	-	-	-	79,600	TSI P-Trak	(Afshari et al., 2005)
Electric stove	-	-	-	111,500	TSI P-Trak	(Afshari et al., 2005)
	-	-	-	218,400	TSI P-Trak	(Afshari et al., 2005)
	-	-	-	116,800	TSI P-Trak	(Afshari et al., 2005)
	1	-	-	17,064	TSI P-Trak	(Weichenthal et al., 2007)
Electric cooking	5	-	-	94,000	TSI 3022A	(Dennekamp et al., 2001)
	6	-	-	111,000	TSI 3022A	(Dennekamp et al., 2001)
	3	-	-	11,000	TSI 3022A	(Dennekamp et al., 2001)
	2	-	-	30,000	TSI 3022A	(Dennekamp et al., 2001)
	2	-	-	24,000	TSI 3022A	(Dennekamp et al., 2001)
	2	-	-	16,000	TSI 3022A	(Dennekamp et al., 2001)
	-	-	-	150,900	TSI P-Trak	(Afshari et al., 2005)
Gas cooking	4	-	-	26,000	TSI 3022A	(Dennekamp et al., 2001)
	6	-	-	146,000	TSI 3022A	(Dennekamp et al., 2001)
	4	-	-	133,000	TSI 3022A	(Dennekamp et al., 2001)
	5	-	-	137,000	TSI 3022A	(Dennekamp et al., 2001)
	2	-	-	98,000	TSI 3022A	(Dennekamp et al., 2001)
	3	-	-	124,000	TSI 3022A	(Dennekamp et al., 2001)
	3	-	-	125,000	TSI 3022A	(Dennekamp et al., 2001)
Cleaning	-	-	-	550	TSI P-Trak	(Afshari et al., 2005)
	-	-	-	7,200	TSI P-Trak	(Afshari et al., 2005)
	-	-	-	21,400	TSI P-Trak	(Afshari et al., 2005)
	-	-	-	38,300	TSI P-Trak	(Afshari et al., 2005)
	1	-	Median:	11,100	TSI 3934, TSI 3022A, TSI 3320	(He et al., 2004)
	3	-	Median:	34,900	TSI 3934, TSI 3022A, TSI 3320	(He et al., 2004)
	5	-	Median:	41,300	TSI 3934, TSI 3022A, TSI 3320	(He et al., 2004)
	17	-	Median:	30,900	TSI 3934, TSI 3022A, TSI 3320	(He et al., 2004)
	1	-	Median:	14,100	TSI 3934, TSI 3022A, TSI 3320	(He et al., 2004)
ETS	-	-	-	213,300	TSI P-Trak	(Afshari et al., 2005)
	6	-	-	26,600	TSI 3934, TSI 3022A, TSI 3320	(He et al., 2004)
Wood burning	-	-	-	17,546	TSI P-Trak	(Weichenthal et al., 2007)
At-work						
Exposure to diesel exhaust	-	31,000	470,000	142,286	TSI P-Trak	(Wheatley and Sadhra, 2004),

4.4. Characterisation of the Study Population

The mean time for continuous UFP measurement for the 22 participants was 44.5 hours and ranged from 4 to 56 hours (Tab. 4.4). Due to implausible data and missing or incomplete diary information, 23.6 out of 936 hours of total sampling time, including 11.7 hours due to implausible sudden changes in UFP levels, were eliminated. The eliminated data included 19 hours of residential-indoor monitoring without active sources, such as, cooking or cleaning; 4 hours of travelling by car, and less than three minutes of cleaning, cooking and working. The overall sampling time available for analysis, therefore, was 912 hours. All 22 participants completely filled in the questionnaire and diary. Due to this high overall availability of data, it can, therefore, be inferred that the study population was highly reliable.

Tab. 4.4 Characteristics of the study population and measurement periods

	N	%		
Men	12	54.5		
Women	10	45.5		
CITY				
Dinslaken	9	40.9		
Duisburg	4	18.2		
Essen	2	9.1		
Mülheim/Ruhr	1	4.5		
Oberhausen	6	27.3		
EMPLOYMENT				
Full-time (≥30 h/week)	7	32		
Part-time (<30 h/week)	6	27		
Not employed	9	41		
	sampling time			
	[h]	[%]		
TIME SPENT IN ME				
Total	912	100		
Residential-indoor	683	74.9		
Residential-outdoor	64	6.8		
At-work	82	7.0		
In-transit	64	6.8		
Other places	19	2.1		
	sampling time		participants	time spent per participant
	[h]	[%]	[n]	[h]
TIME MEASURED FOR SPECIFIC PERIODS				
Cooking	30	3.3	13	2.3
Cleaning	25	2.7	10	2.5
Candle burning	3	0.3	3	1
Car	45	4.9	15	3
Bicycle	12	1.3	4	3
Walking	7	0.8	5	1.4
Periods with open windows	71	7.8	9	7.9
Working	82	9.0	8	10.3
Day (without specific source)	254	27.9	21	12.1
Night (without specific source)	286	31.4	21	16
Residential-outdoor	64	7.0	11	6
Other places	19	2.1	6	3.2
	Mean	Min	Max	
WEATHER CONDITIONS				
Temperature [°C]	15	8	20	
Humidity [%]	70	40.5	96	
Pressure [hPa]	1,014	1,001	1,027	

4.5. Weather Conditions

Weather conditions during sampling were rather inconstant. Detailed readings for humidity, temperature and pressure can be found in Table 4.4 above and Figure 4.4 below. Temperature values during sampling time ranged from 8 to 20°C (mean 15°C). This shift in temperature may have influence on ventilation methods and outdoor activities. A change of humidity may also change particle numbers. Shi and Harrison (1999) showed that higher air humidity resulted in higher UFP readings during diesel engine-induced UFP formation. Since neither of the modelling approaches takes weather conditions into account, it can be assumed that constantly changing air humidity may cause difficulties while comparing measured and modelled data.

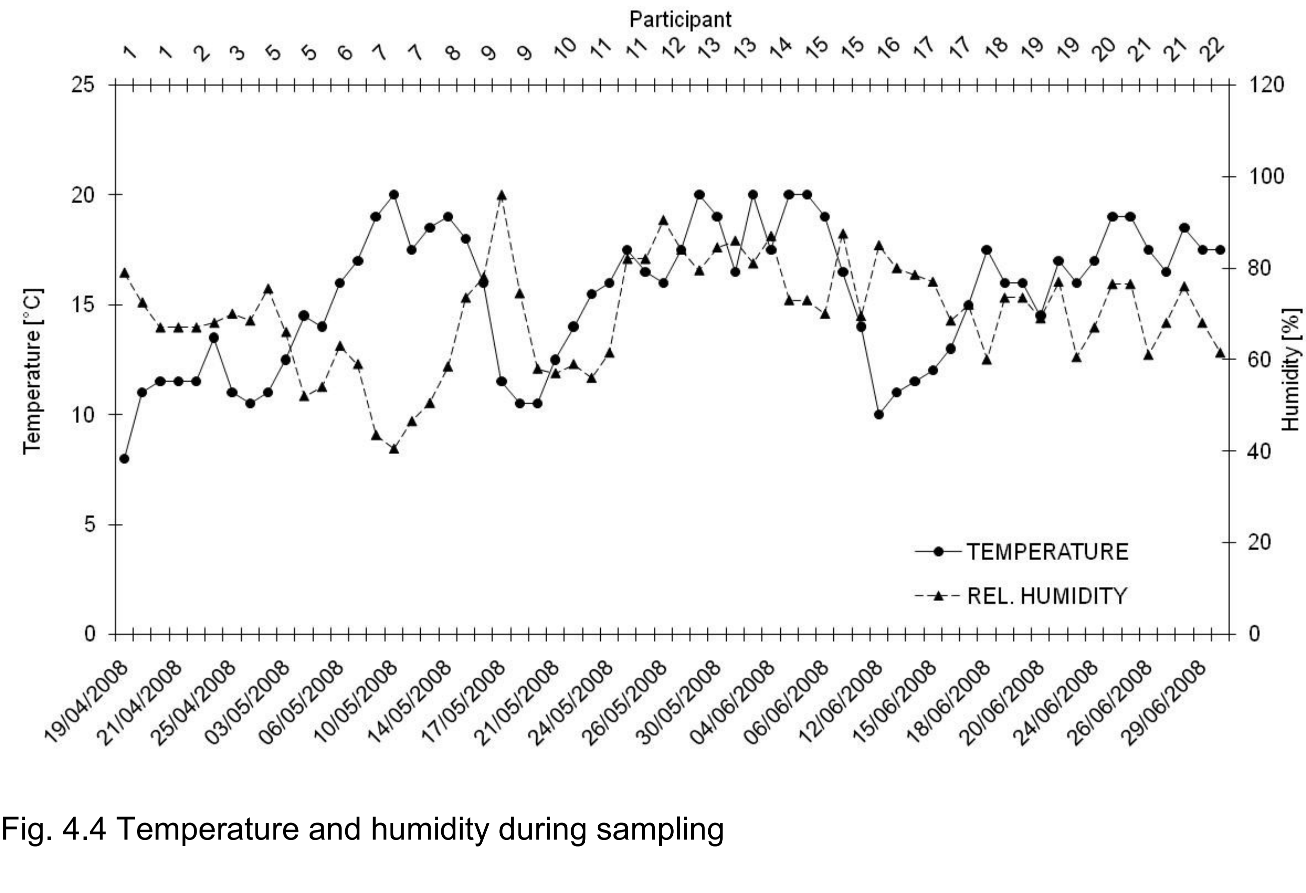

Fig. 4.4 Temperature and humidity during sampling

4.6. Measured UFP Data

Sampling was performed as described in the method section. All measured results for each participant, including exposure levels and time spent in distinct microenvironments, are presented in Table 4.5 below.

Tab. 4.5 Mean UFP concentration and mean time of exposure of participant in each defined microenvironment

Participant	Microenvironments Mean UFP concentration [#/cm³]						Mean time [h]					
	IT	RI	RO	AW	OP	Overall mean	IT	RI	RO	AW	OP	Total
1	8,617	2,825	-	-	131,817	14,061	4.25	39.25	-	-	3.75	47.25
2	-	5,991	-	-	-	5,993	-	41.75	-	-	-	41.75
3	8,269	8,200	-	-	-	8,227	4.75	43.5	-	-	-	48.25
4	13,915	16,683	-	-	12,140	14,654	0.75	45	-	-	2.5	46.75
5	21,283	8,079	-	-	-	8,706	2.5	44	-	-	-	46.5
6	7,355	10,691	12,221	-	-	10,787	2	31	14.75	-	-	47.75
7	-	18,311	19,818	-	32,415	18,931	-	36	13.75	-	1.25	51
8	8,503	822	-	7,255	-	4,141	4.5	15.5	-	20	1	41
9	2,267	1,765	-	-	6,889	2,623	17.5	21	-	17.25	5.5	61.25
10	16,639	5,355	-	6,228	-	6,470	3.25	25.75	-	17.25	-	46.25
11	21,608	15,546	17,137	-	-	16,239	4.25	32	9.75	-	-	46
12	7,259	8,673	7,292	-	-	8,429	1	39.75	7.5	-	-	48.25
13	14,464	9,547	18,016	4,182	-	10,546	9.75	39	2	1.25	-	52
14	10,820	9,678	8,645	-	-	9,648	0.75	55.75	2.25	-	-	58.75
15	8,986	13,187	-	13,729	-	13,138	1.25	37	-	10	-	48.25
16	15,661	9,985	-	-	-	9,933	0.75	43.75	-	-	-	44.5
17	6,500	11,033	7,720	-	-	10,715	1.75	43.75	3	-	-	48.5
18	14,755	11,654	9,998	14,873	26,344	13,329	2.25	31	1	11.75	2.75	48.75
19	15,017	11,626	-	-	14,144	12,384	5.5	36.25	-	-	7.5	49.25
20	12,917	17,727	33,866	10,471	-	13,159	4.5	25	0.25	20.25	-	50
21	12,396	7,877	6,379	6,838	-	7,796	2.25	33.5	3.25	-	-	39
22	2,588	6,186	3,813	-	777	4,933	1.75	36.75	8.25	-	2.75	49.5
mean	11,500	9,611	13,173	9,082	32,075	10,220	3.42	36.19	6	13.96	3.38	48.2

The time range of continuous UFP measurement in the 22 participants was between 4.4 and 56.2 hours. 6 periods had to be discarded due to implausibly high or low data. The overall measurement time was 912 hours. The total measurement time incorporated periods with an identified UFP-emitting source, namely, 30 hours of cooking, 25 hours of cleaning and 45 hours of travelling by car, as well as, 286 hours during the night and 254 hours during the day without a particular source. Overall, 34, 20, 60, 39 and 123 periods of cooking, cleaning, travelling by car, as well as, night and daytime exposure, respectively, were recorded. Due to outliers 23.6 hours of sampling time was eliminated, including 19.4 hours RI without active sources, 4 hours of travelling by car, and less than three minutes of cleaning, cooking and working.

Figure 4.5 shows an example of a complete 48-hour sampling period at 15-minute intervals for participant 12. Diary entries of cleaning and cooking coincide with time periods of high exposure. However, even during time periods without known UFP-emitting sources, peaks of high exposure occurred. During the night, UFP concentration falls continuously until reaching the lowest value in the early morning hours. It can be seen that UFP levels are raised during periods of cooking, and cleaning. Detailed time-activity patterns for each participant can be found in Appendix III.

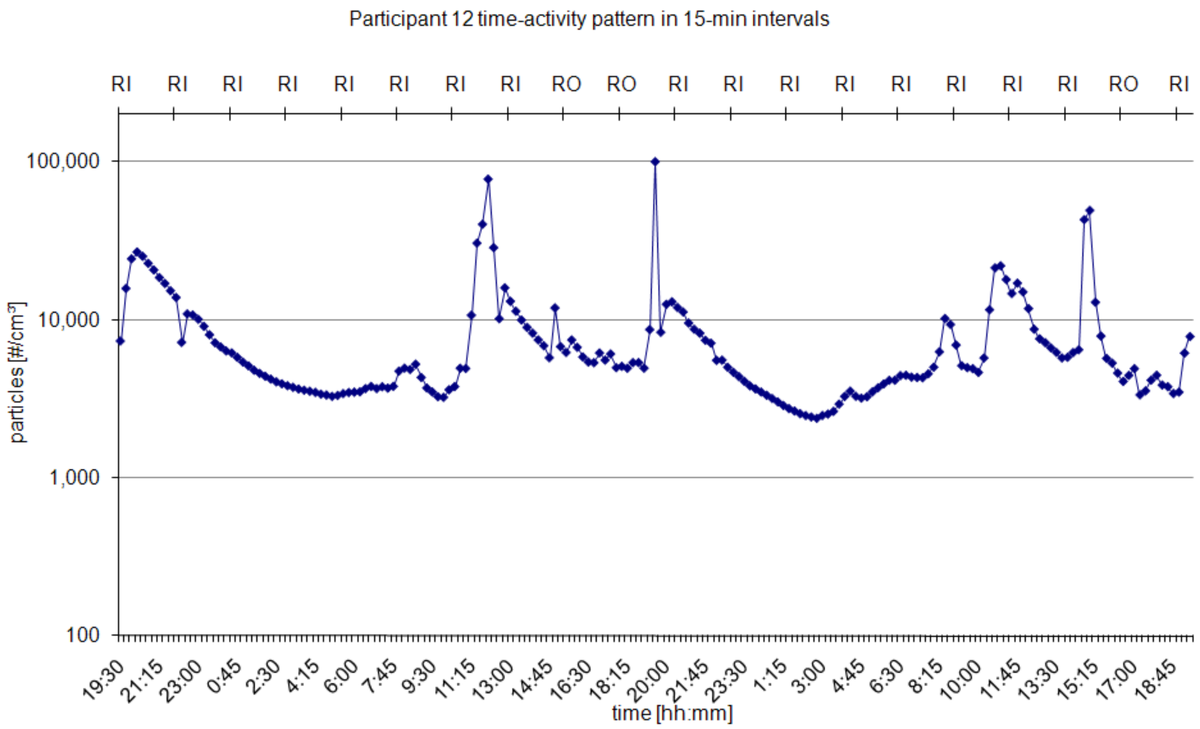

Fig. 4.5 48-hour time-activity pattern for participant 12 with corresponding UFP exposure at 15-minute intervals

Figure 4.6 illustrates the sampling results for each microenvironment and the total sampling time.

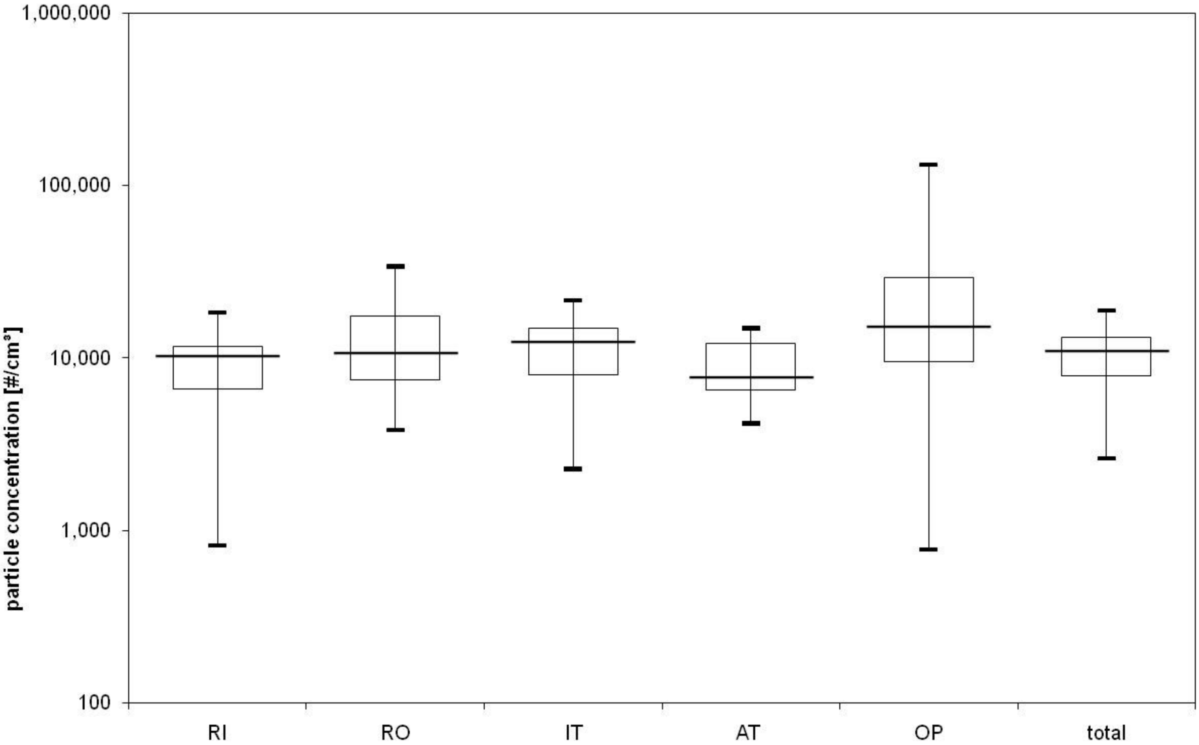

Fig. 4.6 Sampling results for each microenvironment

The highest UFP values during the 912 hours of sampling time were obtained during periods of cooking and in the OP ME - 201,600 #/cm³ and 180,000 #/cm³, respectively (Table 4.6 and Figure 4.7).

Tab. 4.6 Summary data for all recorded exposures in microenvironments and during specific periods including baseline

	Participants [n]	Periods [n]	Sampling Time [h]	Mean duration period [min]	UFP concentration [#/cm³] Mean	Median	Range	Q1	Q3
Residential-indoor (RI)									
Without active sources									
Day and night	21[1]	162	522	281	8,221	8,202	736-91,731	3,847	9,205
Day	21	123	254	122	10,734	7,715	816-91,731	4,928	12,291
Night	18	39	286	440	5,708	4,728	736-49,273	3,059	6,969
Active sources									
Cooking	13	34	30	53	22,556	12,566	988-201,575	5,647	27,878
Cleaning	10	20	25	74	11,004	8,720	1,994-109,082	6,100	14,214
Candles	3	3	3	30	16,780	12,723	3,788-49,522	10,016	18,285
In-transit (IT)									
Car	15	60	45	45	9,613	6,356	982-98,116	2,974	12,067
Bicycle	4	10	12	32	12,178	9,929	4,091-55,661	8,948	12,896
Walking	5	11	7	34	9,667	7,357	1,490-38,968	5,428	11,023
At-work (AW)	8	18	82	273	9,700	8,115	1,552-78,052	5,696	11,561
Other places (OP)	6	74	19	85	26,992	11,560	1,216-179,994	7,555	20,843
Residential-outdoor (RO)	11	236	64	154	12,781	9,897	1,761-91,615	5,678	16,503

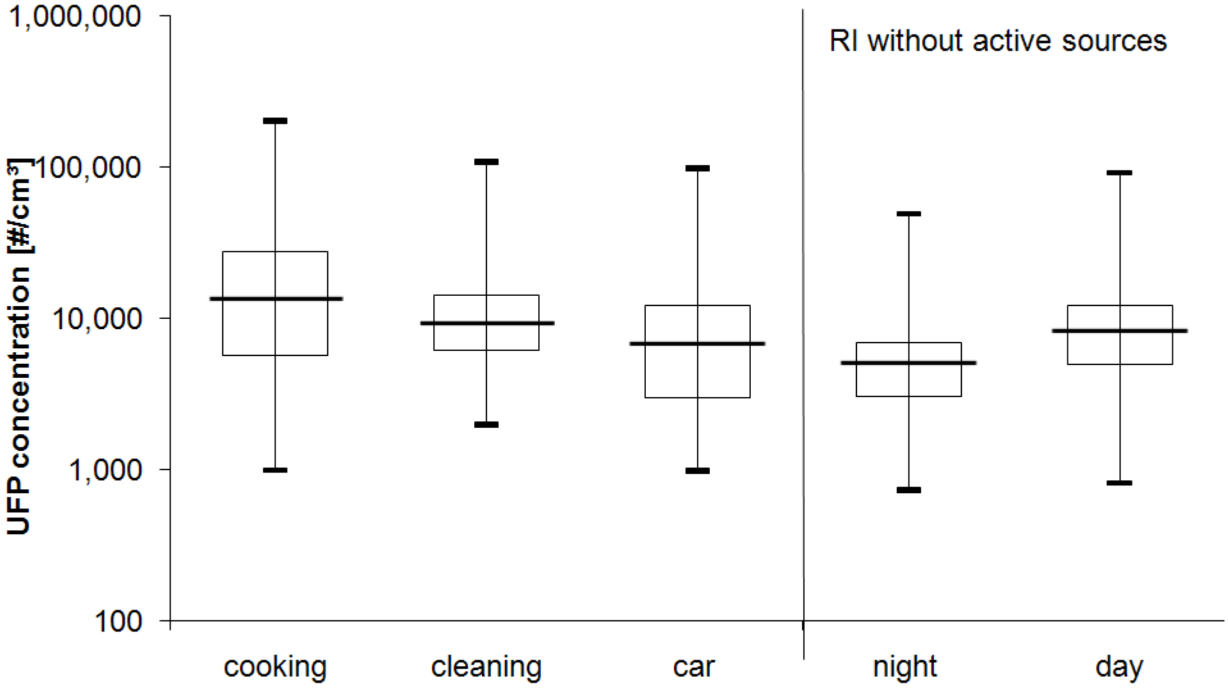

Fig. 4.7 Distribution of UFP concentration for all periods of cooking, cleaning, travelling by car, and ongoing day and night including baseline

The variance of the distribution of 5-minute exposure concentrations is lowest during night and highest whilst cooking (Fig. 4.8). No normal distributions were found, however, the distributions for cooking and cleaning were multimodal, while travelling by car and day and night-time exposure without active sources were unimodal.

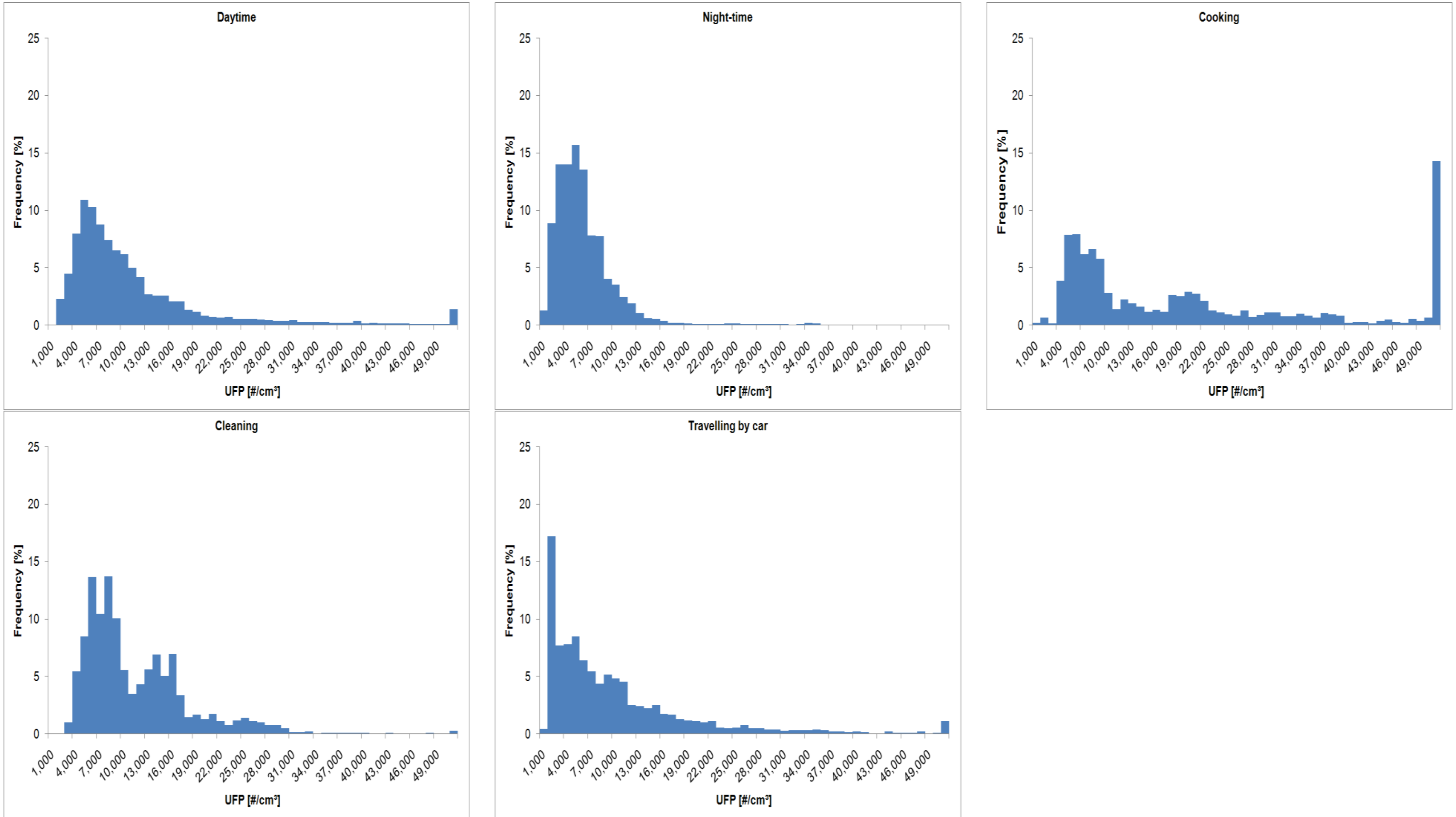

Fig. 4.8 Distribution of UFP concentrations for all microenvironments and sub-environments

4.7. Determination of Baseline Influence

Table 4.7 and Figure 4.9 summarise the increase in UFP exposure during specific periods after subtracting individual baseline values derived from directly preceding time periods when no active source was recorded. While most of the recorded periods actually lead to an increase in UFP exposure, some periods had a negative value. For comparison of in-transit values, the preceding residential-indoor period was used as a baseline.

Tab. 4.7 Difference in UFP measurements before and during specific periods

	Participants	Periods	Sampling time	Mean duration	UFP concentration			
					Mean	SD	Min	Max
	[n]	[n]	[h]	[min]	[#/cm³]			
Residential-indoor (RI)								
Cooking	11	19	16	50	18,701	23,935	-3,350	89,070
Cleaning	10	16	17	66	266	5,578	-12,680	9,860
In-transit (IT)								
Car	13	32	30	52	2,202	12,990	-33,110	30,440

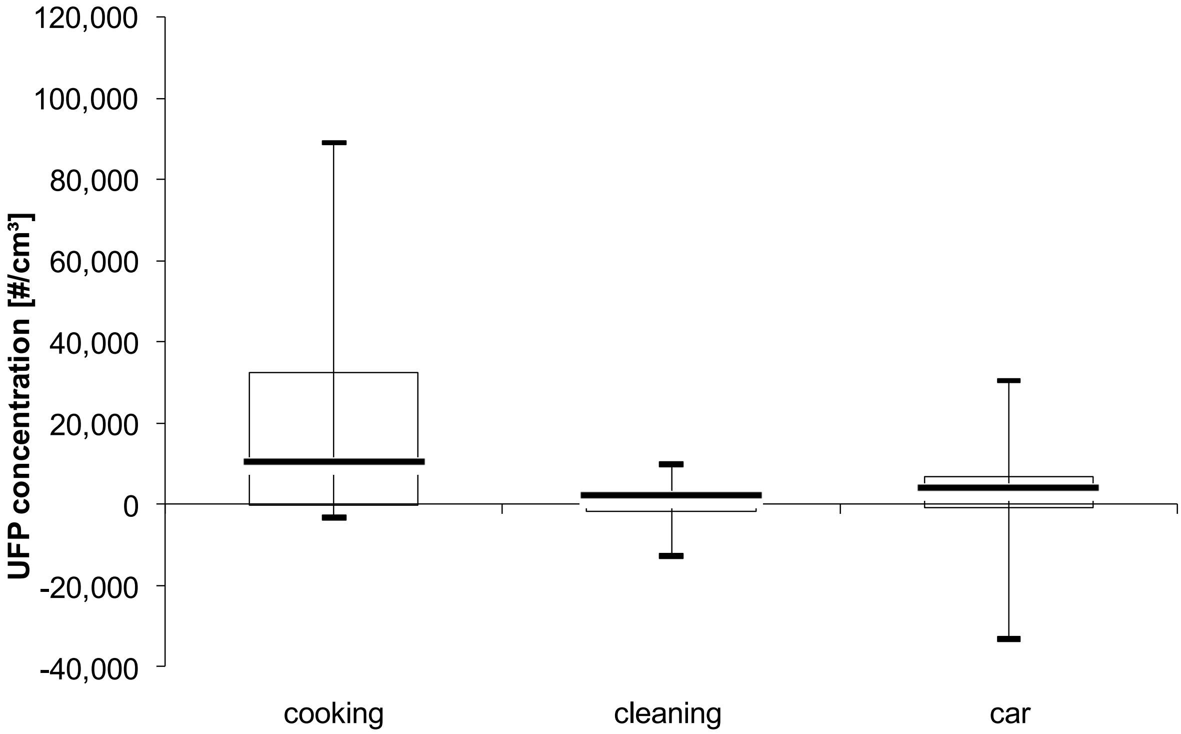

Fig. 4.9 Distribution of UFP concentrations during periods of cooking, cleaning and travelling by car after deduction of corresponding reference period

4.8. Comparison of Measured and Literature Data

Sampled UFP exposure for cooking lies within the range of literature values but shows overall smaller UFP readings. Literature values for cooking were up to 150,900 #/cm³ while sampled data range up to 201,600 #/cm³ due to one very high value. A similar effect could also be seen for cleaning, candle burning, travelling by bicycle and walking. In this pilot study, travelling by car produced higher UFP readings than those obtained from literature (literature up to 20,100 #/cm³, sampled up to 98,100 #/cm³).

4.9. Identification of Main Contributors to UFP Exposure

Table 4.8 and Figure 4.10 present the percentage values for time as well as exposure for specified exposure events for participants who were exposed at least once to a particular event. Participants spent on average 4.8% (0.6-11.3%) of the sampling time cooking. Cooking contributed up to 12% (1.5-26.3%) of the total cumulative exposure, while travelling by car constituted 11.1% (1.3-50.0%) of the sampling time and 13.3%

(2.0-50.2%) of the total UFP exposure. 8.4% (1.8-28.6%) of the sampling time was spent cleaning, which contributed 8.0% (4.0-29.7%) to the total UFP exposure.

Tab. 4.8 Contribution of 22 subjects to the cumulative UFP exposure during the 48-hour sampling time, stratified by specific exposure periods of cooking, cleaning and travelling by car

	Participants exposed at least once to specific source of exposure [n]	Mean CB_{ij} for specific exposure periods [%]	Mean percentage of time spent during specific exposure periods [%]
Cooking	13	12.0	4.8
Cleaning	10	8.0	8.4
Car	15	13.3	11.1

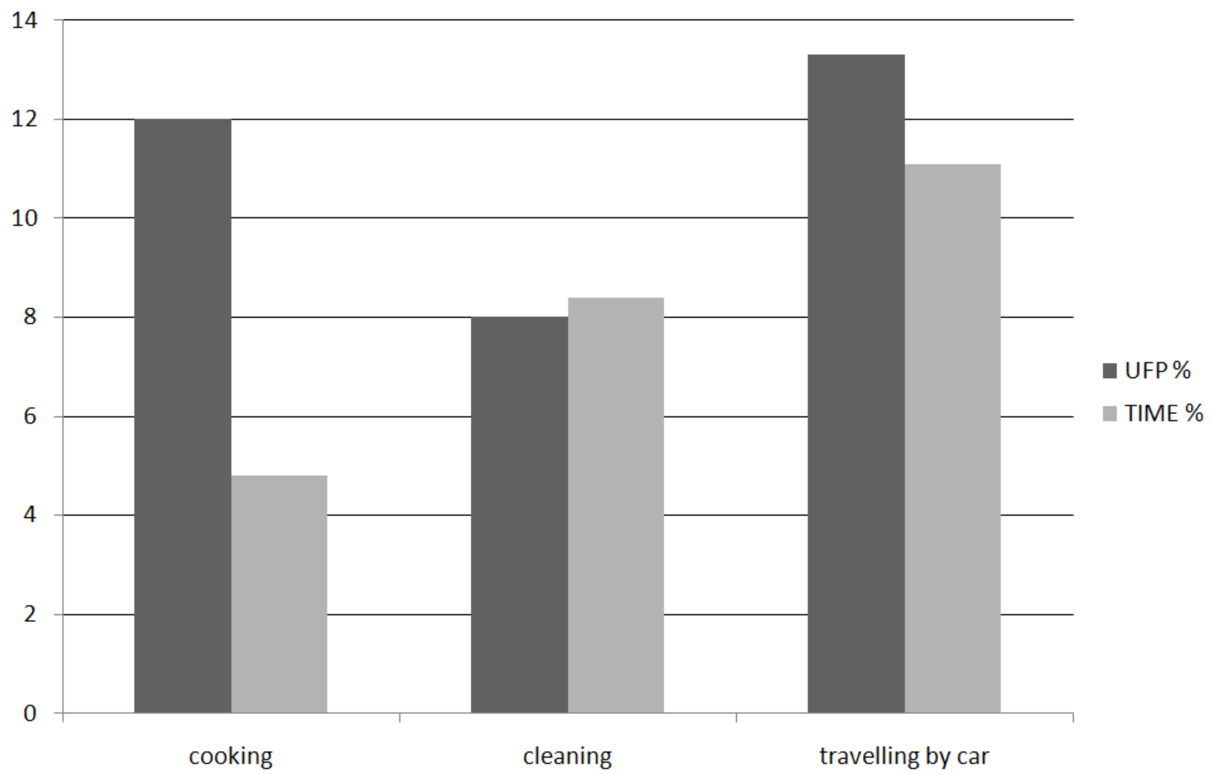

Fig. 4.10 Percentage contribution of specific episodes of cooking, cleaning and travelling by car to UFP exposure and time

4.10. Variability of Sampled Data

Table 4.9 presents data for the variability of the UFP measurements in the various microenvironments and specific exposure periods. A very large overall variability can be seen for all MEs especially for cooking which showed the highest values for $CV_{(tot)}$. Within-subject ($CV_{(i)}$) and within-period ($CV_{(ij)}$) variability are notably smaller, with the lowest values recorded during periods where no active UFP-emitting sources were present.

Tab. 4.9 Variability of sampled data

	$CV_{(tot)}$ for all participants	Mean $CV_{(i)}$ for each participant	Mean $CV_{(ij)}$ for a specific period
Residential-indoor (RI)			
Cooking	1.12	0.67	0.43
Cleaning	0.60	0.41	0.25
Candle burning	0.61	0.18	0.18
Night without active sources	0.84	0.23	0.30
Day without active sources	0.95	0.24	0.59
Day and night without active sources	0.98	0.27	0.41
In-transit (IT)			
Car	1.05	0.53	0.35
Bicycle	0.46	0.30	0.16
Walking	0.67	0.47	0.23
At-work (AW)	0.61	0.42	0.34
Residential-outdoor (RO)	0.77	0.46	0.10
Other places (OP)	1.45	0.45	0.12

4.11. Comparison of Questionnaire and Diary Statements

This section will present the differences found between diary and questionnaire statements. Table 4.10 presents mean statements in hours. A graphical overview for each activity can also be found in Figures 4.11 to 4.17.

Tab. 4.10 Deviations between short-term diary and long-term questionnaire statements [h/d]

	Questionnaire					Diary					
Source/activity	Mean	SD	Min	Max	CI_{95}	Mean	SD	Min	Max	CI_{95}	Deviation
Cooking	1.16	0.85	0.02	4	±0.39	0.34	0.86	0.39	2.88	±0.39	0.82
Cleaning	0.56	0.41	0	1	±0.19	0.28	0.43	0	1.63	±0.20	0.28
Candle burning	0.43	0.59	0	2	±0.27	0.09	0.23	0	1	±0.11	0.34
Time spent at home	15.98	3.16	11	22	±1.44	18.46	4.65	9.78	27.88	±1.84	2.48
Walking	0.76	0.47	0.25	2	±0.21	0.07	0.15	0	0.56	±0.07	0.69
Bicycle	0.64	0.43	0.25	1.25	±0.20	0.07	0.18	0	0.81	±0.08	0.57
Car	1.02	0.38	0.25	1.25	±0.17	0.77	1.86	0	8.63	±0.84	0.25

It can be seen that the mean questionnaire and diary statements differ between 0.25 and 0.82 hours for specified activities or sources, while the difference between the time spent at home is approximately 2.5 hours. The highest deviations were noted for travelling by bicycle and walking. During adverse weather conditions people tend to use the car or public transport rather than riding a bicycle or walking. Furthermore, the TSI handheld CPC 3007 could not be used while jogging, because of the tendency of the device to malfunction when it is jolted.

The following charts highlight the differing statements for all specific sources and the time spent at home. It can be seen from the questionnaires that time was overestimated for all activities and sources except for periods spent at home.

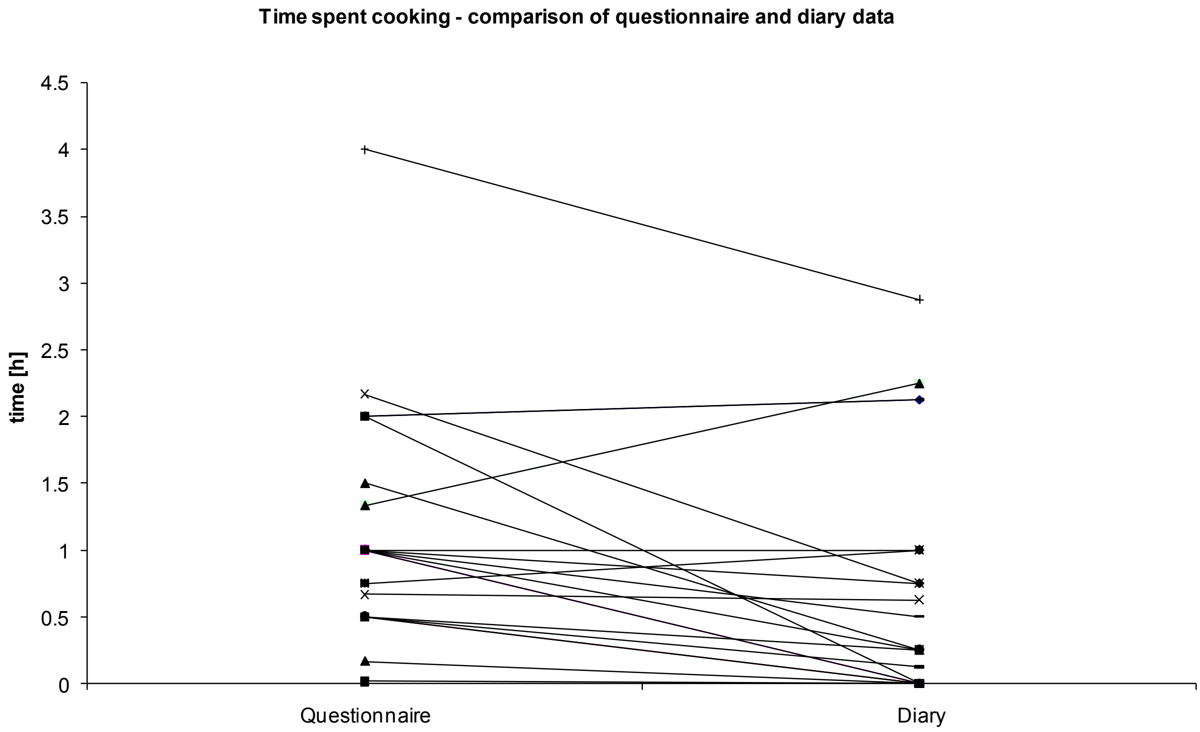

Fig. 4.11 Comparison of questionnaire and diary data for cooking

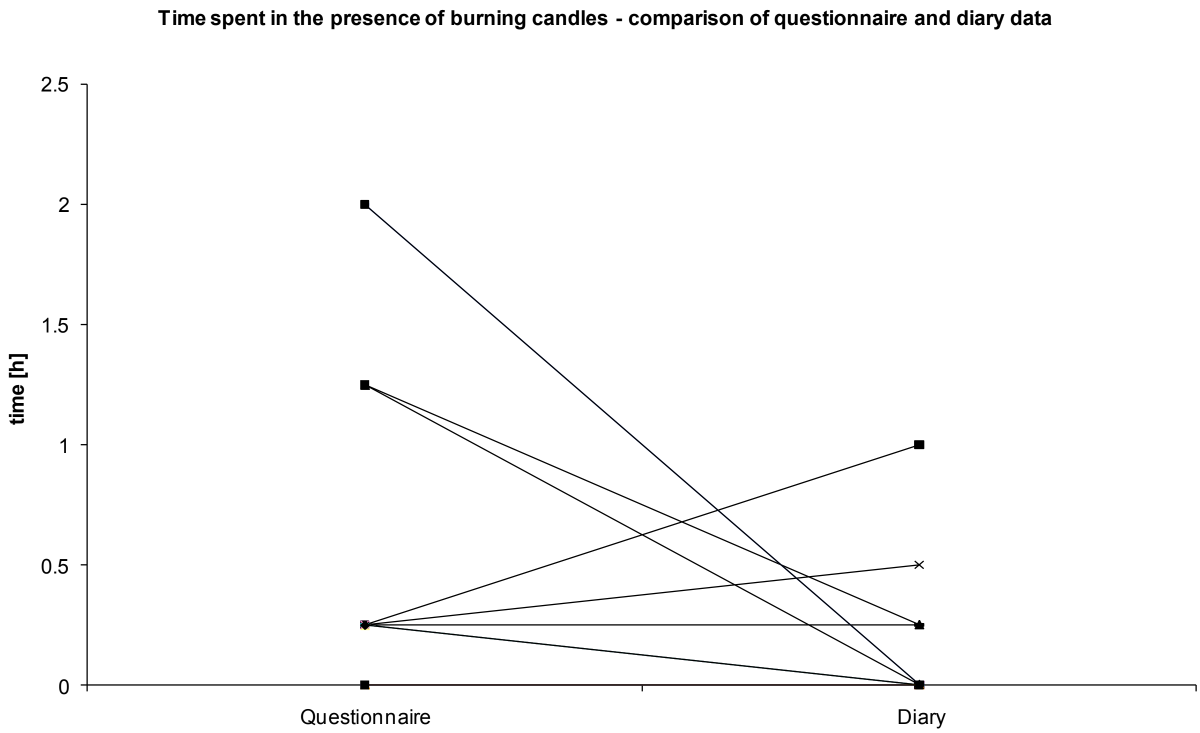

Fig. 4.12 Comparison of questionnaire and diary data for candle burning

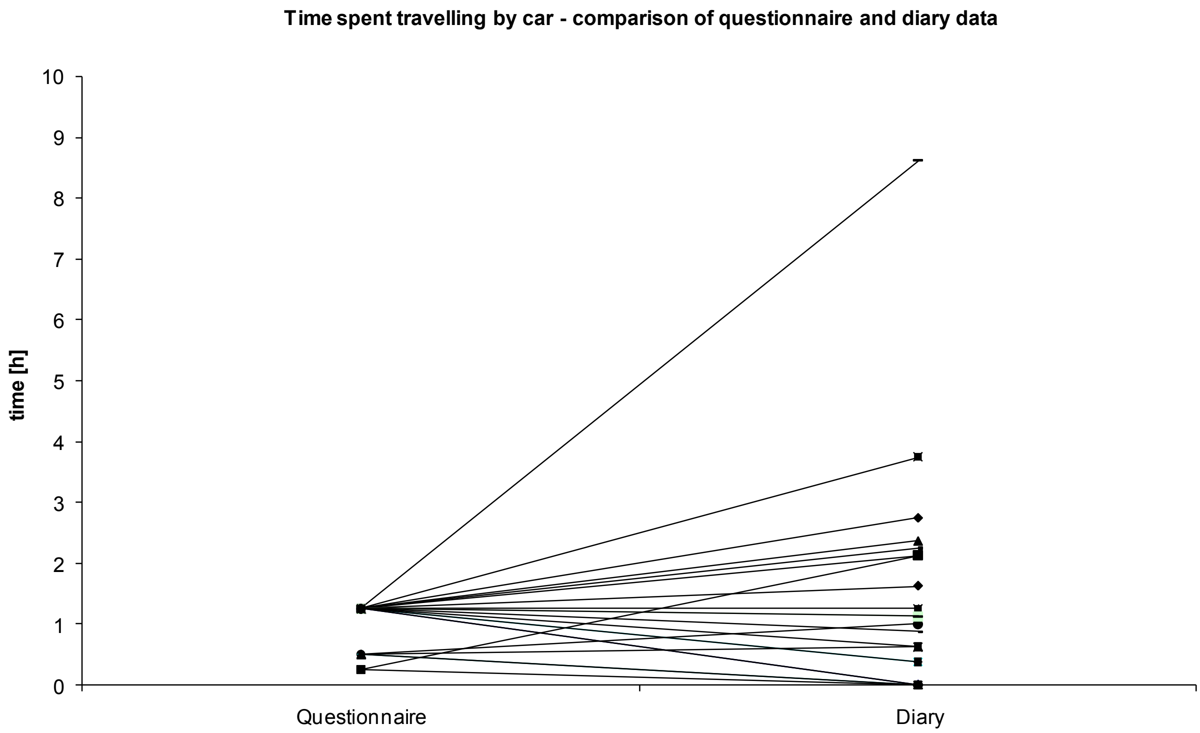

Fig. 4.13 Comparison of questionnaire and diary data for travelling by car

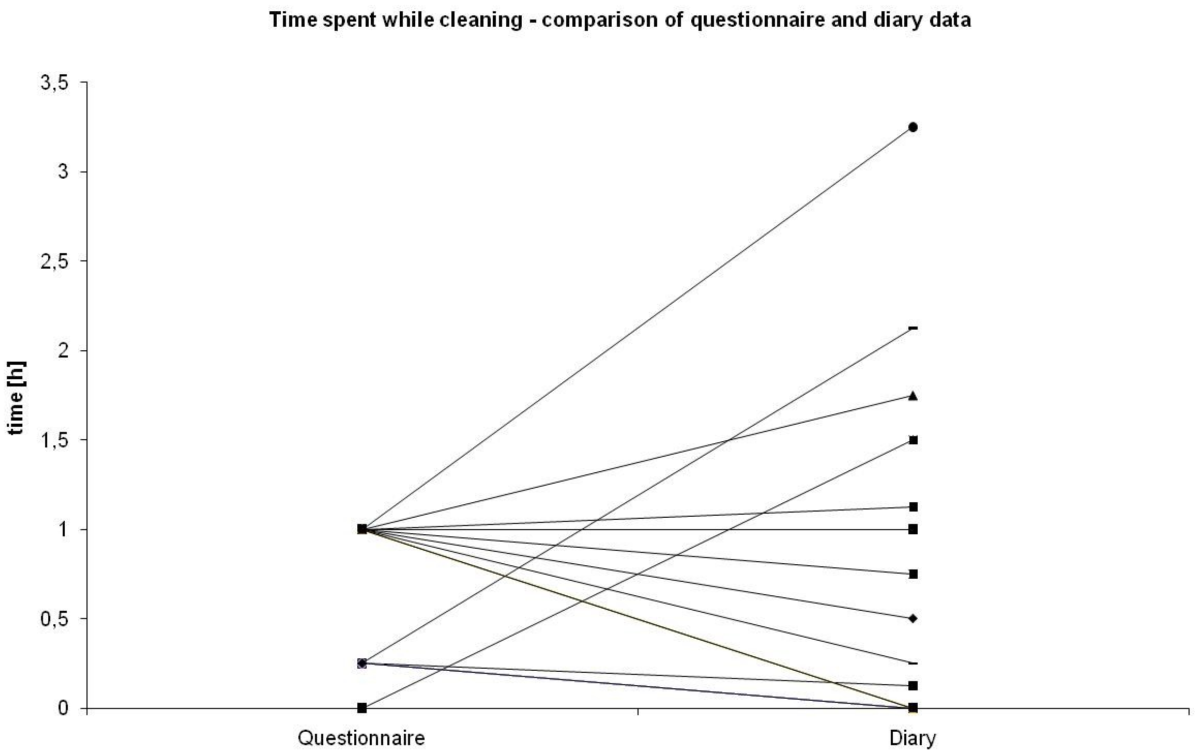

Fig. 4.14 Comparison of questionnaire and diary data for cleaning

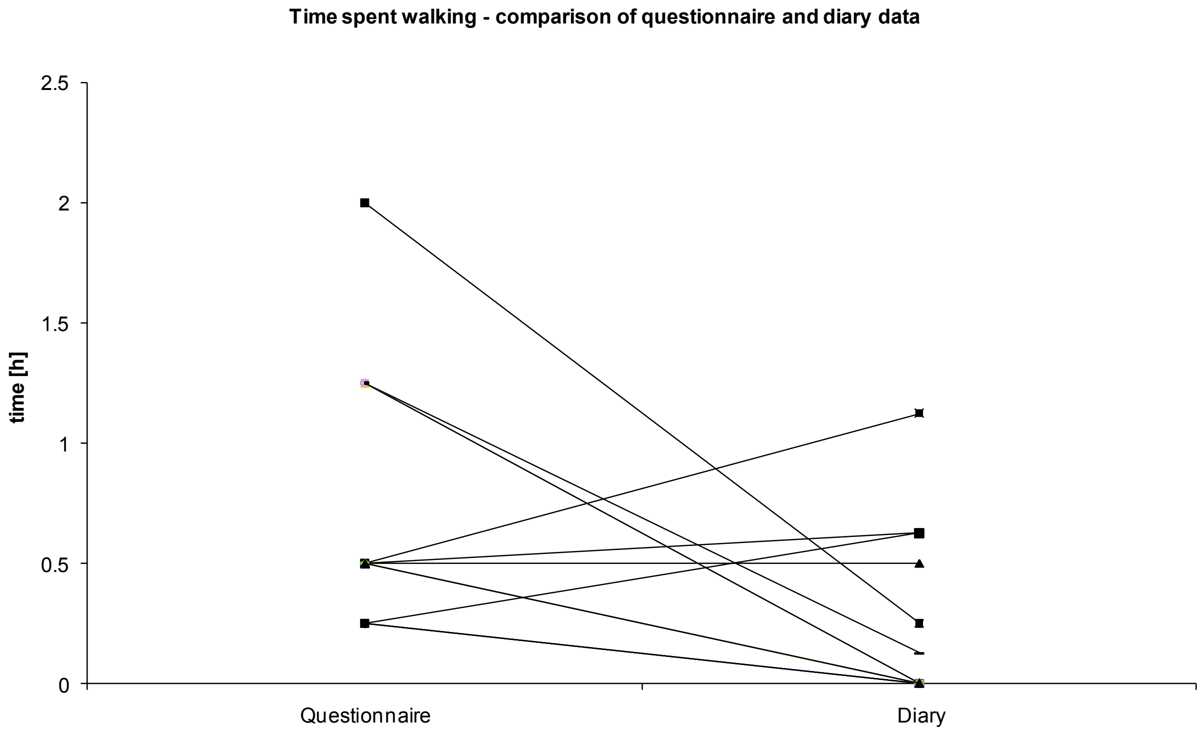

Fig. 4.15 Comparison of questionnaire and diary data for walking

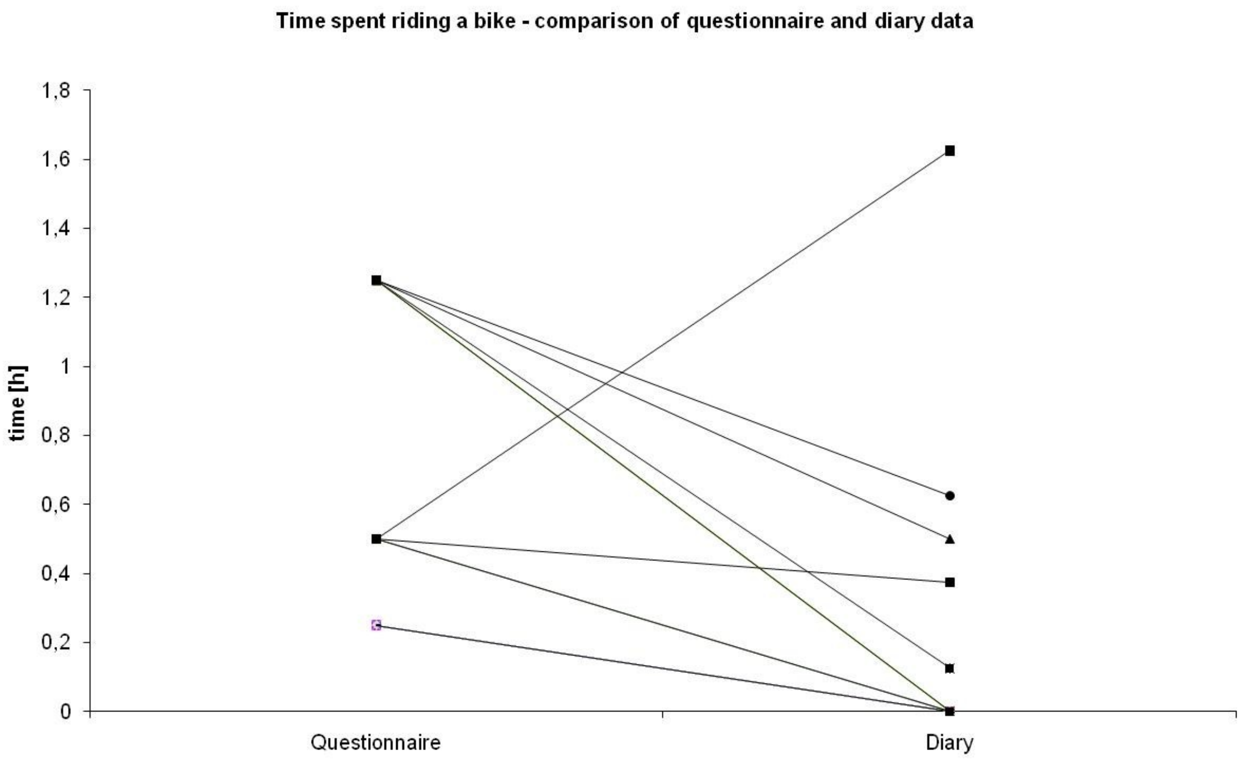

Fig. 4.16 Comparison of questionnaire and diary data for riding a bike

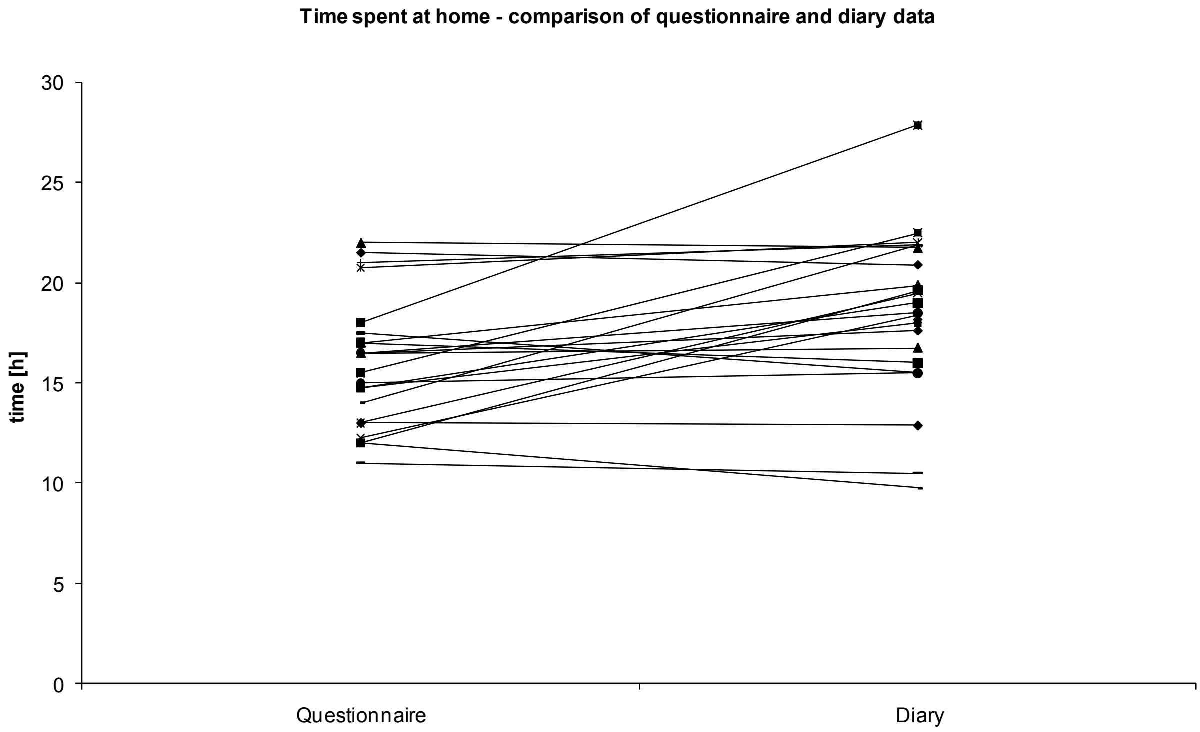

Fig. 4.17 Comparison of questionnaire and diary data for the time spent at home

5. Discussion

5.1. Introduction

In this pilot study personal exposure to UFP was sampled continuously over a period of 48 hours under real life conditions. It can be seen that personal UFP exposure as measured by a handheld particle counter varies to a great extent depending on personal activities and location. A stay in microenvironments with specific active sources, which were defined on the basis of their expected influence on exposure, generally coincided with an increase in UFP exposure. However, the range of values for specific MEs and periods was wide and varied considerably between and within participants. Cooking and travelling by car were identified as important contributors to overall personal UFP exposure.

5.2. Sampling Data within Different Microenvironments and Periods

The highest UFP concentrations were identified within the residential-indoor and in-transit microenvironment when a specific UFP-emitting source was present. On the other hand, exposure within the indoor home microenvironment was generally low when no specific source was active (mean 8,200 #/cm³). Within the residential-indoor microenvironment, the highest increase in UFP concentration was found during periods of cooking (maximum 201,600 #/cm³). These high UFP exposures while cooking are a result of combustion processes. Although participants in this study only used electrical appliances for cooking, it is important to differentiate between different methods of cooking, such as, frying or baking, as well as the energy source used, such as, gas or electricity, as these different methods of cooking have a varying effect on the amount of UFP released (Dennekamp et al., 2001; He et al., 2004). Values obtained while cooking are comparable with those found in the literature - the literature mean exposure ranging between 11,000 and 150,900 #/cm³ (Dennekamp et al., 2001; He et al., 2004).

Candle burning as well as active and passive smoking are combustion processes similar to cooking which lead to increased indoor UFP concentrations. It was found that candle burning raised UFP levels up to 49,500 #/cm³. Literature reports UFP levels from 69,000 to 241,500 #/cm³ for candle smoke (Afshari et al., 2005; Sun et al., 2007). Due to sampling results for only two participants and five periods, no further conclusions could be drawn for this source.

Cleaning is another activity associated with high UFP concentrations up to 109,100 #/cm³. He et al. (2004) and Afshari et al. (2005) found lower values of up to 41,300 #/cm³, depending on the type of cleaning process. Studies concentrating on cleaning-related exposure found vacuuming to be the most important source (up to 41,300 #/cm³), while other activities, such as washing, ironing and dusting resulted in lower concentrations of up to 30,900 #/cm³ (He et al., 2004; Szymczak et al., 2007). Raised UFP levels during cleaning are a result of re-suspension processes (Abt et al., 2000; He et al., 2004).

Within the in-transit microenvironment it was found that travelling by car produced the highest increases in UFP exposure up to 98,100 #/cm³ in comparison with other modes of transportation, such as walking (up to 39,000 #/cm³) or riding a bicycle (max 55,700 #/cm³). This was also found by Cattaneo et al. (2009) who compared walking, travelling by bus and by car. Concentrations for travelling by car found in literature ranged from 1,900 to 107,000 #/cm³ (Hill and Gooch, 2007; Cattaneo et al., 2009). These high readings may be due to the production of high concentrations of UFP-emitting combustion processes, such as, burning of fuel, that enter the cabin while driving. Exposure while travelling by car is also dependent on the surrounding environment, the ventilation system used, the car's filter system, as well as, whether windows are open or closed (Rudell et al., 1999; Long et al., 2002; Cattaneo et al., 2009).

5.3. Variability of Sampled Data

The UFP concentrations varied considerably not only between different microenvironments and specific exposure situations but also within the specific period itself. Sources of variability of UFP concentration between this study and prior studies, as well as, between measurements in this study include: (a) the experimental conditions, (b) the definition of specific exposure situations, (c) diary precision and time allocation, (d) unrecorded UFP-emitting sources and (e) external factors, such as, weather conditions, background shifts in UFP concentration and local UFP sources. Variations between sampled and literature values result from the different experimental conditions, such as, different sampling devices and the degree of control over the experimental setting. During this pilot study, UFP concentrations were sampled under real life conditions. These differ in several ways from experimental conditions where type and duration of the UFP source can be controlled by the investigator.

Probably the most important source of variability within this study is the definition of specific exposure situations. UFP-emitting activities can be strictly defined during experimental studies, however, the time allocated by the participant to specific activities in a real life situation is very subjective. In this study, for example, one was unable to differentiate between the time where the cooking appliance was actually turned on and the other periods involved in the preparation of food. For future modelling purposes, it is important to differentiate between actual cooking itself, such as, frying, and the cleaning and preparation steps. The distance between the measuring device and the UFP-emitting source also significantly influences the observed number concentrations due to the rapid formation of agglomerates.

Furthermore, UFP concentrations were allocated according to the 15-minute diary statements and not according to actual changes in UFP concentration. During the 15-minute period recorded in the vicinity of a particular UFP-emitting source, the participant may have actually changed location or activity. The participant could, for example, have stopped cooking and moved on to a different activity or location with no UFP-emitting source. In this case, the diary describes a period of cooking, but the sampled data actually shows a change in UFP concentration. A reduction from 15- to 5-minute intervals may help to correlate sampled data with diary statements; however, the degree of detail a participant can record in a questionnaire is limited. In addition, there might be a discrepancy between the TSI's internal clock and the participant's own. In case of retrospective filling of the diary, timing errors of 15 minutes or more are likely.

Other sources which have not been noted in the diary, such as industrialisation, high population density and proximity to high traffic volumes, may have influenced sampling results. Activities of neighbours or housemates were not assessed but they may also have lead to UFP emissions contributing to the monitored UFP concentrations. Even simple movement of a person within a microenvironment raises UFP counts (Abt et al., 2000). This may have altered concentration readings in this study. Furthermore, the diary was not designed to gather information on UFP-emitting sources that were not being actively used by the participant. In order to develop a personal exposure model, the diary should also carefully assess activities in the participant's immediate vicinity.

Changing weather conditions may be a further reason for variability. In this study with only relatively short sampling times, however, the relationship between relative humidity, temperature and UFP concentration could not be identified. Furthermore, the majority of the sampling time was spent indoors, therefore, it can be assumed that changes in weather conditions might not have had such a major influence on the sampled UFP concentration.

Higher variability for periods of cooking, cleaning and travelling by car across all participants were found (coefficient of variation 0.6-1.12) in contrast to other periods without active emitting sources. The CVs of all periods of performing a specific activity within the same individual were 0.41 to 0.67. The lowest variation of exposure was seen within a subject and within a specific period (CV 0.25-0.43). Daytime UFP concentrations when no specific UFP source was active have a higher variability (mean 10,700 #/cm^3, CV 0.59) than readings taken during the night (mean 5,700 #/cm^3, CV 0.30), indicating possible higher background shifts during the day.

5.4. Contributors to Overall Personal UFP Exposure

The contribution of the residential-indoor microenvironment to the total UFP exposure during the day was significantly higher than that at night. The lower UFP concentrations at night may be due to lower traffic counts as well as to inactive UFP-emitting sources, while high daytime concentrations may be due to active UFP-emitting sources, such as, higher traffic counts or sources that were overlooked by the study, such as, activities performed by housemates. Furthermore, combustion-related processes could be identified as important contributors to personal UFP exposure. It can also be shown that cooking is an important specific contributor to total cumulative UFP exposure within the residential-indoor microenvironment. While only 4.8% of the total time was spent cooking, this activity contributed 12.0% to the cumulative exposure during the 48-hour sampling period. In comparison to other modes of transportation, namely walking or riding a bicycle, travelling by car was also an important source of UFP exposure within the in-transit microenvironment (time spent 11.1%; UFP contribution 13.3%).

A similar comparison cannot be made between different cleaning methods as the study was not designed to differentiate between them. It was found, however, that 8.4% of the time was spent cleaning and that this contributed to 8.0% of the total cumulative UFP exposure. Future studies should differentiate between the various cooking and cleaning methods, such as, sweeping, dusting or washing. Other sources and activities, namely walking or candle burning, may also be important, however, further information is needed for modelling purposes. These sources could not be investigated in this study due to the lack of sufficient data.

5.5. Limitations and Strengths of Data Collection

The small number of participants N=22 in this pilot study is an important limitation. All findings are based on a small dataset of observations in microenvironments and during specific exposure situations. Furthermore, this study could only make use of one mobile TSI sampler. Therefore, background periods were limited to periods without active UFP-emitting sources. A second UFP sampler could have been utilised in order to obtain background concentrations during the entire sampling time. Due to the small number of observations in specific exposure situations, such as, candle burning, riding a bicycle, or being exposed to environmental tobacco smoke, this study had to concentrate on only a few UFP sources, such as cooking, cleaning and travelling by car where longer sampling times could be recorded. Additional sources, such as, candle burning, bicycle riding or exposure to environmental tobacco smoke, which could possibly be important in other populations, could not be studied in more detail.

The strength of this study is that UFP concentrations could be sampled under real life conditions while most other studies were conducted under technical or laboratory conditions. Furthermore, most studies focussed on the assessment of personal $PM_{2.5}$ exposure. This study was able to explore the extent to which sampled UFP concentrations under experimental conditions correlate with real life situations.

5.6. Comparison of Questionnaire and Diary Data

A comparison of questionnaire and diary data showed that time periods were clearly overestimated in the questionnaire. This may be due to several reasons:

(i) The questionnaire might have been too general without offering specific choices. Participants who never travel by bicycle, for example, had to state that they use a bicycle ‘never or occasionally’. This statement was interpreted as a maximum use of 15 minutes per day. Furthermore, the questionnaire was designed for collection of data on an ordinary weekday. Sampling, however, also took place during weekends, when participants’ behaviour might possibly have been different from that on a typical weekend. Variation in behaviour may also occur during leisure time. This may also influence the choice of transportation, as well as indoor activities, such as, cleaning or cooking.

(ii) The participants might not have accurately assessed their actual habits while filling in the questionnaire. This could further explain over- or underestimation of the time spent performing a particular activity.

(iii) The presence of the TSI sampler might have triggered a certain curiosity and encouraged participants to perform certain activities when they would not normally do so. One example is the burning of candles in order to provoke a reaction to the emitted particles by the TSI sampler. This might also have induced further discrepancies between diary and questionnaire statements. It is clearly understood that differences between diary and questionnaire statements may have a major influence on the outcome of the model. The model is, therefore, directly dependent on the participants’ statements. Comparison between the modelled and the sampled data is, furthermore, complicated by variations between these two sets of data.

5.7. Modelling of Personal UFP Exposure

It can be seen that the mathematical modelling approach is based on numerous assumptions, such as, seasonal variations in habits like heating and ventilation, as well as, by periods of stay within the vicinity of UFP-emitting sources like heaters or ETS. These assumptions were made due to the lack of sufficient data and may, therefore, be one of the weaknesses of this study.

The model's input data from literature does not reflect 'real-life' situations and data obtained is, furthermore, highly dependent on the ambient concentration and the surrounding environment. These present further shortcomings in the study. In order to develop an accurate personal model for UFP exposure, a higher level of detail is required.

5.8. Conclusion

This study was able to identify cooking and travelling by car as important exposure situations for the cumulative personal exposure to UFP. However, it may be assumed that without further detail on specific UFP-emitting sources, the use of sampled data is only partly suited for the development of personal exposure models based on a microenvironmental approach. High inter- and intra-personal variability of UFP exposure in specific exposure situations necessitates a detailed questionnaire assessment of modifying factors.

5.9. Outlook

Personal exposure models are powerful tools in epidemiology. The pilot study clearly indicated that sampled data can be allocated to defined microenvironments and specific exposure periods. The developed model shows weak correlation between sampled and modelled data but can be used as a matrix for the design of future projects with large study populations and altered data assessment methods, such as, more detailed questionnaires and diary data. The sampled data was well within range of literature findings but also showed high variability when compared to literature data. This model could also be used to extend and to possibly fill any gaps in the existing literature database.

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Appendix I

Layout of German questionnaire

Der nachfolgende Fragebogen dient zur Abschätzung der persönlichen Feinstaubbelastung. Die hier erhobenen Daten werden vertraulich behandelt und für Dritte unzugänglich aufbewahrt.

Bei Rückfragen stehe ich Ihnen gerne zur Verfügung unter _____/______ oder ____/______ oder per Email _________@______.de .

Name	…………………………………………		
Alter	……………………………………Jahre		
Geschlecht	m [ ]	w [ ]	verweigert [ ]

1. Schätzen Sie bitte wie weit Sie täglich zu Fuß gehen, entweder zur Arbeit, zum Einkaufen, zur Entspannung etc.?

Weniger als 1 Kilometer	[ ]
1 bis 2 Kilometer	[ ]
Mehr als 2 Kilometer	[ ]
Verweigert	[ ]
Weiß nicht	[ ]

2. Halten Sie sich bei der Arbeit häufiger in Räumen auf, in denen geraucht wird?

Ja	[ ]
Nein	[ ]
Verweigert	[ ]
Weiß nicht	[ ]

3. Halten Sie sich zu Hause häufiger in Räumen auf, in denen geraucht wird?

Ja	[ ]
Nein	[ ]
Verweigert	[ ]
Weiß nicht	[ ]

4. Halten Sie sich tagsüber oder abends häufiger in Räumen auf, in denen geraucht wird? (nicht zu Hause und nicht bei der Arbeit)

Ja	[ ]
Nein	[ ]
Verweigert	[ ]
Weiß nicht	[ ]

5. Wie viele Personen rauchen in Ihrem Haushalt (Sie selbst eingerechnet)?

Anzahl: ………………

Verweigert = 88
Weiß nicht = 99

6. Wie weit liegt Ihre Wohnung (Luftlinie) von einer verkehrsreichen Straße (Berufs- oder Durchgangsverkehr) entfernt?

Weniger als 10 Meter [ ]
10 Meter bis 50 Meter [ ]
Mehr als 50 Meter [ ]

7. Ist die Straße, in der Sie wohnen, beidseits begrenzt von geschlossenen Häuserfronten („Straßenschlucht")?

Ja [ ]
Nein [ ]

8. Wenn nein, was trifft zu? Mehrere Antworten möglich!

Beidseits Häuserfront mit Lücken (d.h. Abstände zum nächsten Haus z.B. Hofeinfahrt) [ ]
Mindestens auf einer Seite freistehende Häuser mit Gärten [ ]
Wohnhaus steht an einer Kreuzung [ ]
Anderes [ ]

9. Auf welcher Etage liegt Ihr hauptsächlich genutzter Wohnraum?

Erdgeschoss oder Souterrain [ ]
1. Etage [ ]
2. Etage [ ]
Höher [ ]

10. Wie lange halten Sie sich ungefähr in Ihrer Wohnung auf (inkl. Nachts)?

	Werktags	Sonntags
Sommer:	………………… Stunden / Tag	………………. Stunden/ Tag
Winter :	………………… Stunden / Tag	………………..Stunden/ Tag

11. Wenn Sie sich im Wohn- oder Schlafzimmer aufhalten, haben Sie dann die Fenster gewöhnlich (die meiste Zeit) geöffnet oder geschlossen?

	Hauptsächliche genutzter Wohnraum		Schlafraum	
	offen	geschlossen	offen	geschlossen
Sommer	[ ]	[ ]	[ ]	[ ]
Winter	[ ]	[ ]	[ ]	[ ]

12. Wie oft wird in Ihrem hauptsächlich genutzten Wohnraum gründlich gelüftet (Fenster weit auf oder zwei gegenüberliegende Fenster „auf Kipp")?

Sommer: …………. mal / Tag oder Dauerlüftung [ ]

Winter: …………. mal / Tag oder Dauerlüftung [ ]

13. Wie lange halten Sie sich durchschnittlich in verrauchten Räumen auf (bei der Arbeit und Privat)? Wenn Sie sich gar nicht in verrauchten Räumen aufhalten, schreiben Sie bitte „0“.

Sommer: ………. Stunden und ……… Minuten pro Tag
Winter: ………..Stunden und ……….Minuten pro Tag

14. Werden in Ihrem Haushalt einzeln zu bedienende Heizöfen (z.B. Kachelöfen, oder Kamin, Gasbrenner, Ölbrenner) benutzt?

Nein [ ]
Ja [ ]

14a. Wenn ja, in welchen Räumen? Mehrfachantworten möglich!

	Holz	Kohle	Öl	Gas	Strom
Küche	[ ]	[ ]	[ ]	[ ]	[ ]
Wohnzimmer	[ ]	[ ]	[ ]	[ ]	[ ]
Schlafzimmer	[ ]	[ ]	[ ]	[ ]	[ ]
Sonstige Räume	[ ]	[ ]	[ ]	[ ]	[ ]

15. Zu welcher Straße liegen die folgenden Räume Ihrer Wohnung? Wenn ein Raum Fenster zu mehreren Straßen hat, wählen Sie bitte die lauteste Straße.

	Hauptsächlich genutzter Wohnraum	Schlafraum
Zur Autobahn	[ ]	[ ]
Zur Hauptverkehrsstraße	[ ]	[ ]
Zur Nebenstraße	[ ]	[ ]
Zum Innenhof mit offener Bebauung	[ ]	[ ]
Zum Innenhof mit geschl. Bebauung	[ ]	[ ]
Zu einer Grünanlage	[ ]	[ ]

16. Welche Beschaffenheit haben Ihre Fenster? Haben Sie …. ?

	Hauptsächlich genutzter Wohnraum	Schlafraum
Schallschutzfenster	[ ]	[ ]
Doppelfenster mit Isovergl.	[ ]	[ ]
Einfachfenster	[ ]	[ ]
Sonstige Fenster	[ ]	[ ]

Wenn ja, welche? Bitte eintragen:……………………….

17. Wie lange halten Sie sich durchschnittlich in Räumen auf, in denen zu dieser Zeit am Herd gekocht oder gebacken wird? (eigenes Kochen mit eingeschlossen)

……………. Stunden und …………… Minuten pro Tag

Mit welchem Herd wird dann meistens gekocht bzw. gebacken?

Gasherd	[ ]
Elektrischer Herd	[ ]
Anderer	[ ] nämlich ……………

Ist dann meistens eine Dunstabzugshaube mit Verbindung zur Außenluft an?

Nein	[ ]
Ja	[ ]
Weiß nicht	[ ]

18. Wie lange halten Sie sich pro Tag in Räumen auf in denen zu gleichen Zeit sauber gemacht wird (z.B. Staubsaugen, Fegen, Staubwischen, Betten machen), entweder von Ihnen oder anderen?

Gar nicht	Weniger als 30 min	30 min bis höchstens 2 Stunden	Mehr als 2 Stunden
[ ]	[ ]	[ ]	[ ]

19. Wie lange halten Sie sich in Räumen auf in denen Kerzen, Teelichter, Duftöllampen oder ähnliches brennen?

Gar nicht	Weniger als 30 min	30 min bis höchstens 2 Stunden	Mehr als 2 Stunden
[ ]	[ ]	[ ]	[ ]

20. Geben Sie bitte an, ob und wie lange Sie folgende Transportmittel benutzen. Wenn Sie ein Transportmittel nicht benutzen, kreuzen Sie bitte „Gar nicht“ an.

	Gar nicht oder nur gelegentlich	Weniger als 30 Minuten pro Tag	30 Minuten bis höchstens 2 Stunden pro Tag	Mehr als 2 Stunden pro Tag
Auto, LKW (auch Beifahrer)	[ ]	[ ]	[ ]	[ ]
Motorrad (auch Beifahrer)	[ ]	[ ]	[ ]	[ ]
Bus/ Straßenbahn	[ ]	[ ]	[ ]	[ ]
U-Bahn	[ ]	[ ]	[ ]	[ ]
Deutsche Bahn	[ ]	[ ]	[ ]	[ ]
Zu Fuß gehen (in der Stadt)	[ ]	[ ]	[ ]	[ ]
Fahrrad (in der Stadt)	[ ]	[ ]	[ ]	[ ]

21. Wenn Sie im Auto fahren (auch als Beifahrer), wie wird dann meistens gelüftet?
Mehrfachantworten möglich!

	Fenster geöffnet	Lüftung Klimaanlage	Gar nicht
Sommer	[ ]	[ ]	[ ]
Winter	[ ]	[ ]	[ ]

22. Nur für Berufstätige:
Wie weit liegt Ihr persönlicher Arbeitsplatz (Luftlinie) von einer verkehrsreichen Straße (Berufs- oder Durchgangsverkehr) entfernt?

Weniger als 10 Meter	[ ]
10 Meter bis 50 Meter	[ ]
Mehr als 50 Meter	[ ]

23. Nur für Berufstätige:
Welche Tätigkeiten werden von Ihnen oder den Kollegen in Ihrer Nähe regelmäßig durchgeführt?

	Ja	Nein
Schweißen	[ ]	[ ]
Gießen	[ ]	[ ]
Löten	[ ]	[ ]
Andere Metallverarbeitung	[ ]	[ ]
Schüttgüter-Umschlag	[ ]	[ ]
Arbeiten Sie in einer Halle mit Kfz-Verkehr (z.B, Lagerhalle, Bushof, Eisstadion, Garage, Kfz-Werkstatt)?	[ ]	[ ]

Vielen Dank fürs Ausfüllen

Appendix II

Diary Layout

Datum:			AUFENTHALTSORT														
Name:			Unterwegs							zu Hause					am Arbeitsplatz		
Stunde	Minute	Kurze Beschreibung der Aktivität und des Aufenthaltsortes falls abweichend	Laufen	Motorrad	Auto / LKW	Bus / Staßenbahn	U-Bahn	Zug	Fahrrad	Aufenthalt in verrauchten Räumen	Kochen	Putzen	Kerzen brennen	Lüften	Umgang mit Schüttgütern	Metallverarbeitung	Aufenthalt in verrauchten Räumen
0	0		O	O	O	O	O	O	O	O	O	O	O	O	O	O	O
	15		O	O	O	O	O	O	O	O	O	O	O	O	O	O	O
	30		O	O	O	O	O	O	O	O	O	O	O	O	O	O	O
	45		O	O	O	O	O	O	O	O	O	O	O	O	O	O	O
1	0		O	O	O	O	O	O	O	O	O	O	O	O	O	O	O
	15		O	O	O	O	O	O	O	O	O	O	O	O	O	O	O
	30		O	O	O	O	O	O	O	O	O	O	O	O	O	O	O
	45		O	O	O	O	O	O	O	O	O	O	O	O	O	O	O
2	0		O	O	O	O	O	O	O	O	O	O	O	O	O	O	O
	15		O	O	O	O	O	O	O	O	O	O	O	O	O	O	O

Appendix III

48-hour Exposure Overview for Each Participant

Participant 1

Tab. III.1 Time-pattern participant 1

Age: 26 ME	Sex: male Time [h]	Activity/location	Time [h]
IT	4.25	Car	4.25
RI	39.25	Sleeping	19.75[1]
		Home	19.5
OP	3.75	Other/ETS	3.75

[1]measured 18.5h

Tab. III.2 Activity-pattern participant 1

ME	Particle concentration [#/cm³]				Activity/location	Particle concentration [#/cm³]		
	Mean	Min	Max	SD		Mean	Min	Max
IT	8,617	5313	13,614	2,346	Car	8,617	5,313	13,614
RI	2,825	1065	5,877	1,257	Sleeping	1,909	1,065	3,041
					Home	3,274	1,304	5,877
OP	131,817	81,628	188,842	38,277	Other/ETS	131,817	81,628	188,842

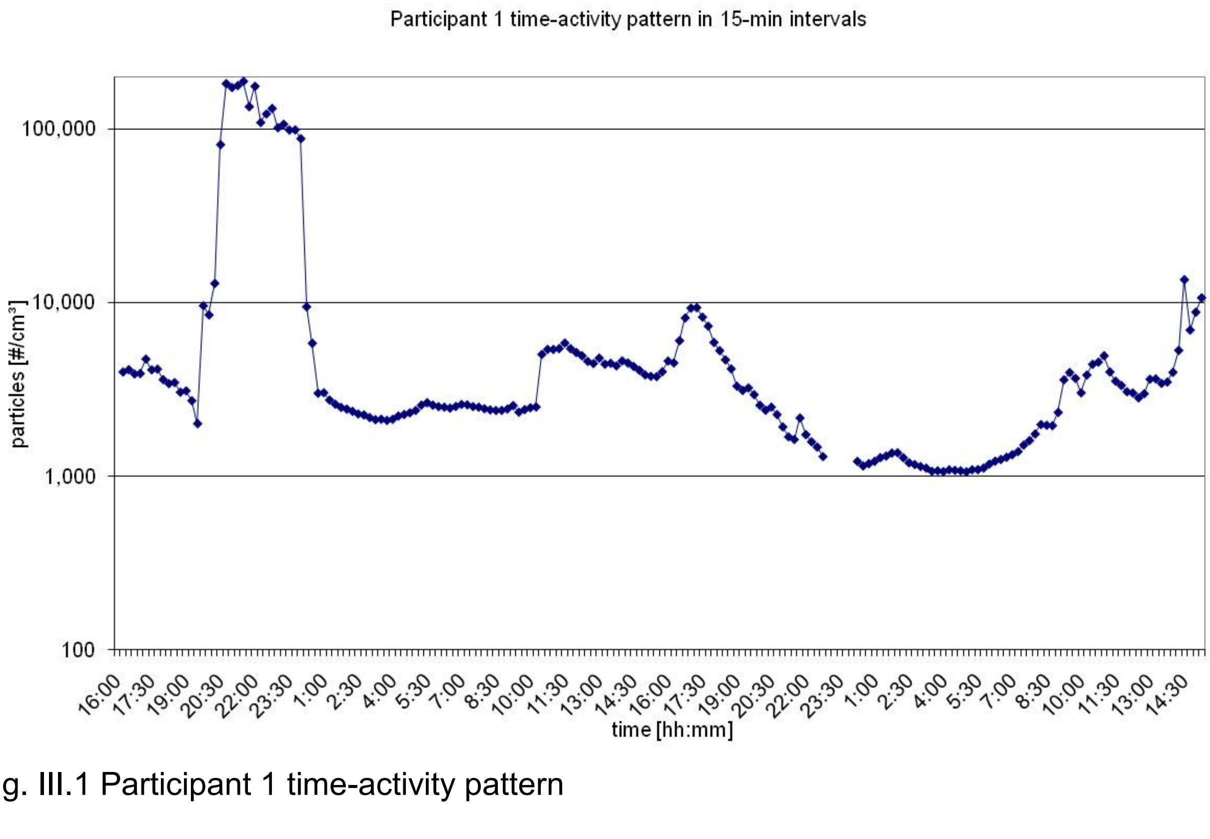

Fig. III.1 Participant 1 time-activity pattern

Participant 2

Tab. III.3 Time-pattern participant 2

Age: 62 ME	Sex: male Time [h]	Activity/location	Time [h]
RI	41.75	Sleeping	18.25
		Home	15.75
		Ventilating	4.25
		Cleaning	0.25
		Cooking/ventilating	1.25
		Cooking	2

Tab. III.4 Activity-pattern participant 2

ME	Particle concentration [#/cm³] Mean	Min	Max	SD	Activity/location	Particle concentration [#/cm³] Mean	Min	Max
RI	5,991	1,196	69,332	9,589	Sleeping	2,008	1,196	6,041
					Home	5,755	2,038	48,440
					Cooking	15,582	3,483	52,757
					Cleaning	3,706	3,706	3,706
					Ventilating	5,191	1,218	7,763
					Cooking/ventilating	16,516	7,466	33,505

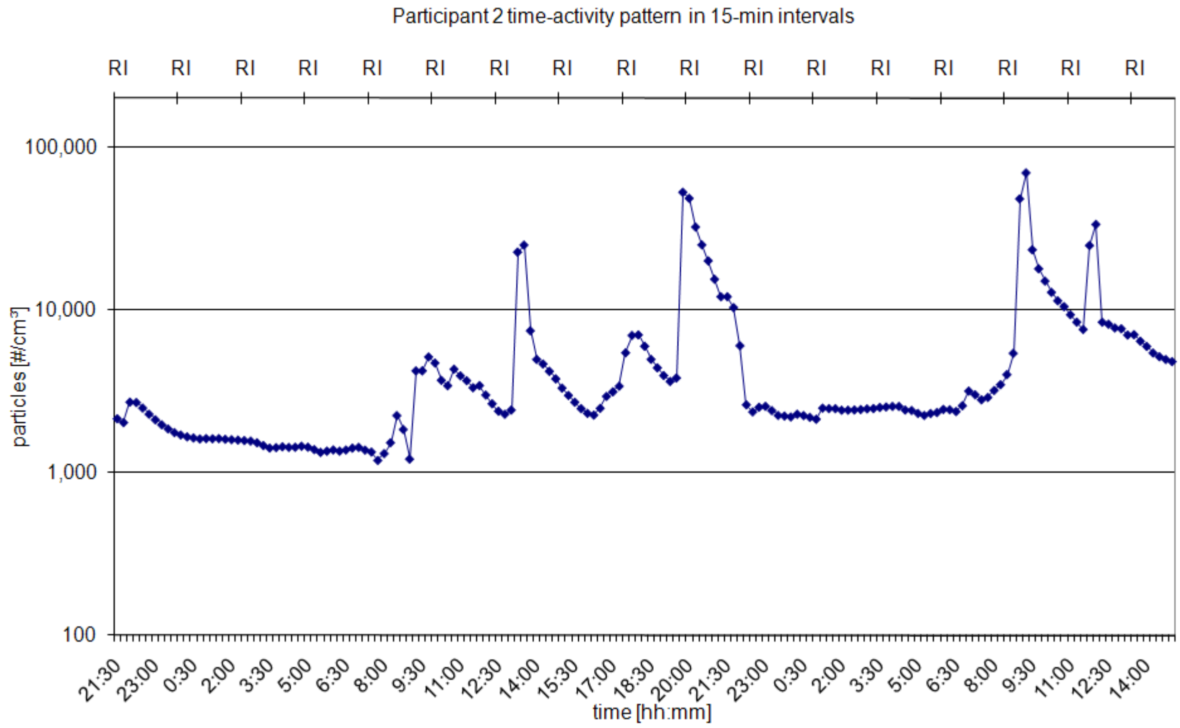

Fig. III.2 Participant 2 time-activity pattern

Participant 3

Tab. III.5 Time-pattern participant 3

Age: 54	Sex: female		
ME	Time [h]	Activity/location	Time [h]
IT	4.75[1]	Car	4.75
RI	43.5	Sleeping	17[2]
		Home	23.25[3]
		Cooking	0.5[2]
		Cleaning	1
		Candles	0.25[2]
		Ventilating	1[3]
		Cooking/ventilating	0.5[2]

[1] measured 1.75h
[2] measured 0h
[3] measured 0.25h

Tab. III.6 activity-pattern participant 3

ME	Particle concentration [#/cm³]				Activity/location	Particle concentration [#/cm³]		
	Mean	Min	Max	SD		Mean	Min	Max
IT	8,269	4,411	14,510	3,621	Car	8,269	4,411	14,510
RI	8,200	5,880	9,495	1,230	Sleeping	-	-	-
					Home	7,421	5,880	8,962
					Cooking	-	-	-
					Cleaning	8,567	7,304	9,495
					Ventilating	8,289	8,289	8,289
					Candles	-	-	-
					Cooking/ventilating	-	-	-

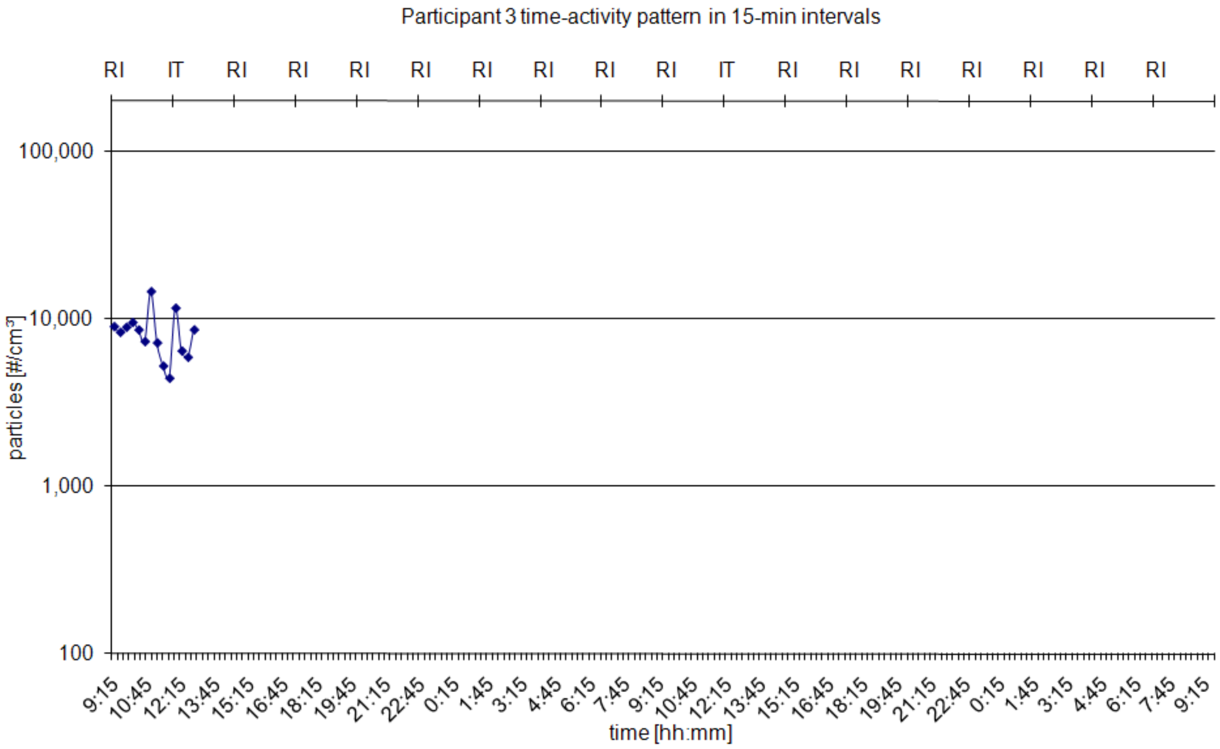

Fig. III.3 Participant 3 time-activity pattern

Participant 4

Tab. III.7 time-pattern participant 4

Age: 68 ME	Sex: male Time [h]	Activity/location	Time [h]
IT	0.75	Car	0.75
RI	45[1]	Home	44.5
		Cooking	0.5
OP	2.5	Other	2.5

[1]measured 7.5h

Tab. III.8 activity-pattern participant 4

ME	Particle concentration [#/cm³] Mean	Min	Max	SD	Activity/location	Particle concentration [#/cm³] Mean	Min	Max
IT	13,915	11,493	17,772	3,376	Car	13,915	11,493	17,772
RI	16,683	7,408	36,489	7,645	Home	13,583	7,408	29,288
					Cooking	30,799	25,110	36,489
OP	12,140	5,177	17,402	4,404	Other	12,140	5,177	17,402

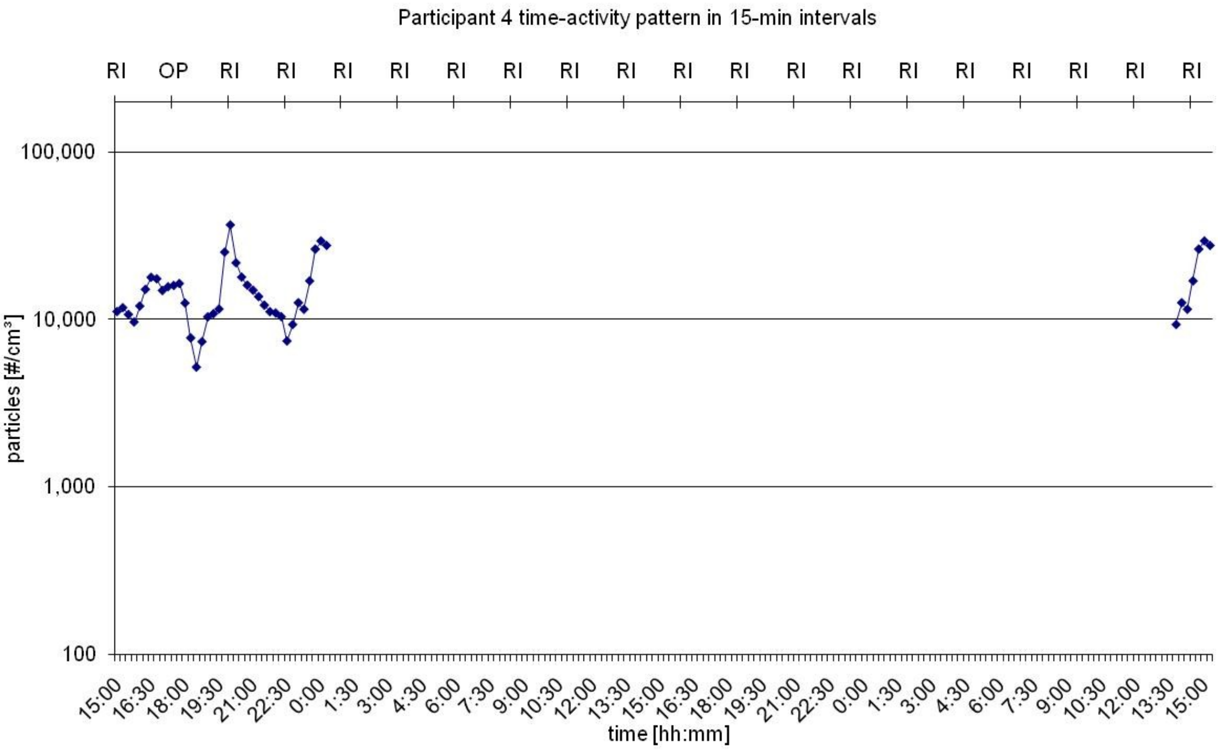

Fig. III.4 Participant 4 time-activity pattern

Participant 5

Tab. III.9 time-pattern participant 5

Age: 84	Sex: male		
ME	Time [h]	Activity/location	Time [h]
IT	2.5	Car	2.5
RI	44	Sleeping	20
		Home	24

Tab. III.10 activity-pattern participant 5

ME	Particle concentration [#/cm³]				Activity/location	Particle concentration [#/cm³]		
	Mean	Min	Max	SD		Mean	Min	Max
IT	21,283	14,658	30,238	5,572	Car	21,283	14,658	30,238
RI	8,079	2,580	48,888	7,067	Sleeping	4,990	2,580	48,888
					Home	13,285	3,638	37,705

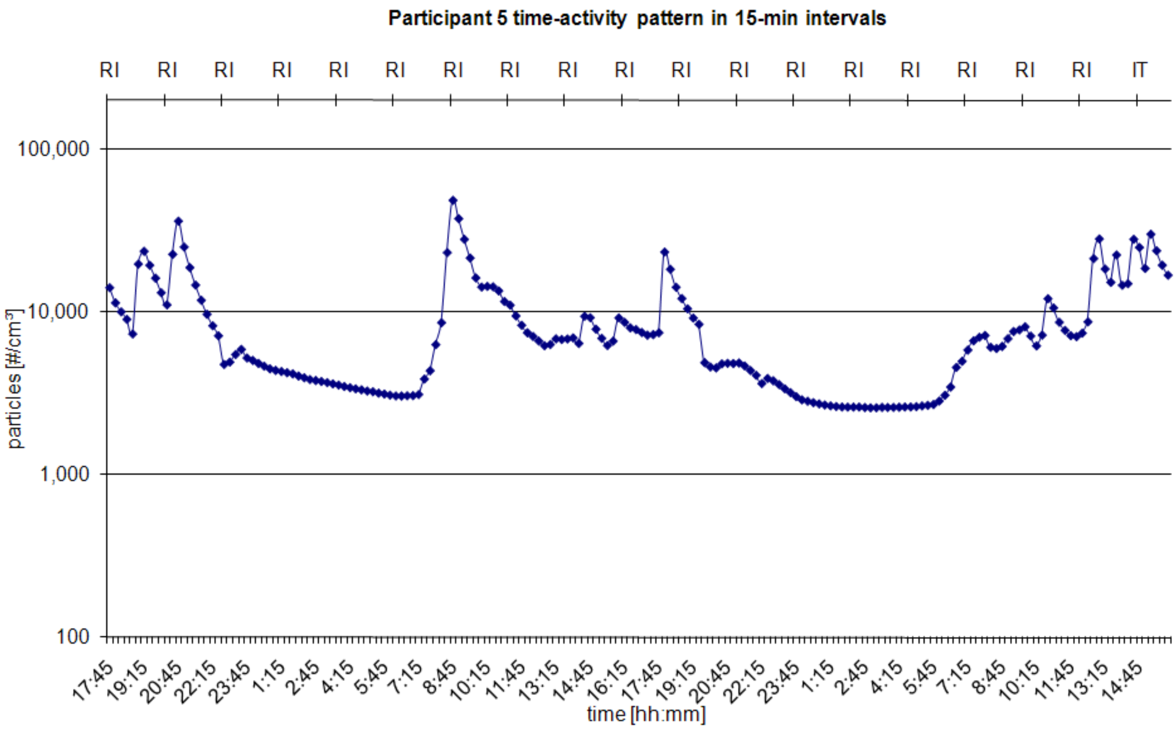

Fig. III.5 Participant 5 time-activity pattern

Participant 6

Tab. III.11 time-pattern participant 6

Age: 57	Sex: male		
ME	Time [h]	Activity/location	Time [h]
IT	2	Car	2
RI	31	Sleeping	16.5
		Home	14.5
RO	14.75	Garden	14.75

Tab. III.12 activity-pattern participant 6

ME	Particle concentration [#/cm³]				Activity/location	Particle concentration [#/cm³]		
	Mean	Min	Max	SD		Mean	Min	Max
IT	7,355	5,980	94,438	1,203	Car	7,355	5,980	94,438
RI	10,691	3,869	54,031	8,087	Sleeping	7,224	3,869	14,283
					Home	14,927	5,629	54,031
RO	12,221	4,457	31,227	4,816	Garden	12,220	4,457	31,227

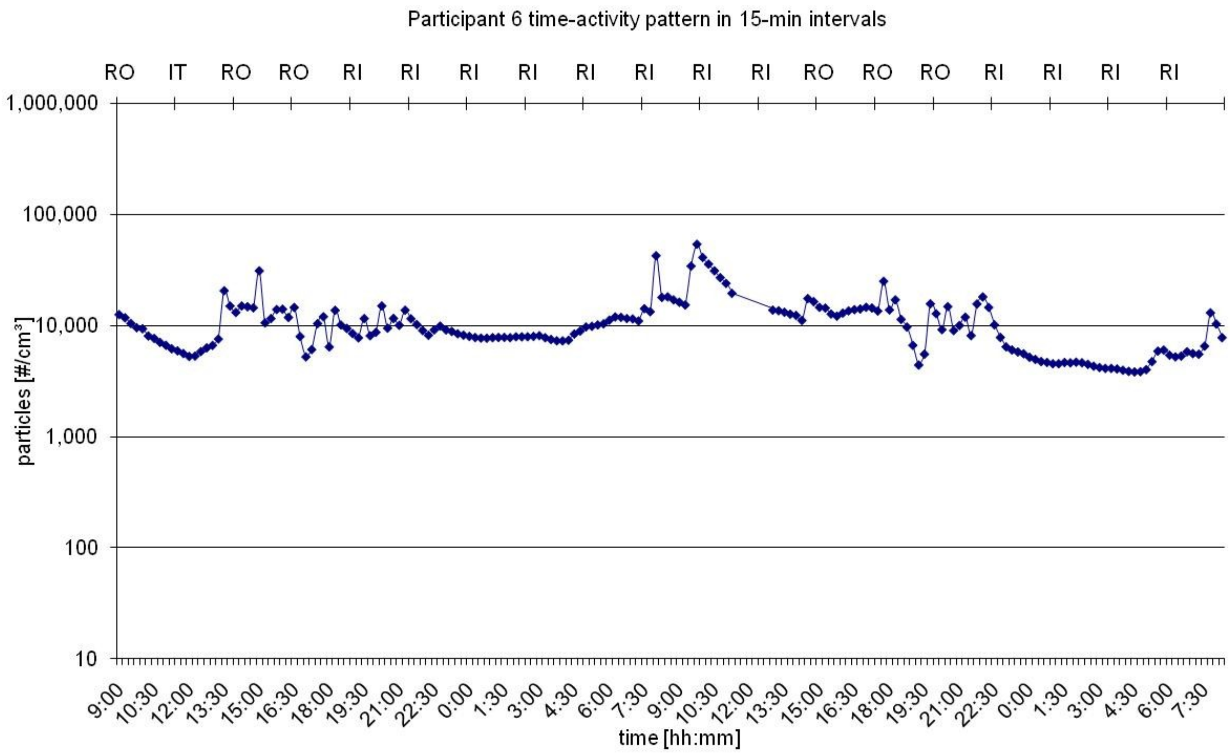

Fig. III.6 Participant 6 time-activity pattern

Participant 7

Tab. III.13 time-pattern participant 7

Age: 55	Sex: female		
ME	Time [h]	Activity/location	Time [h]
RI	36	Sleeping	19.25
		Cleaning	6.5
		Home	4.75
		Cooking	5.75
RO	13.75	Balcony	2
		Garden	11.75
OP	1.25	Other	1.25

Tab. III.14 activity-pattern participant 7

ME	Particle concentration [#/cm³]				Activity/location	Particle concentration [#/cm³]		
	Mean	Min	Max	SD		Mean	Min	Max
RI	18,311	4,381	109,148	14,685	Sleeping	16,015	5,414	55,559
					Home	30,768	10,119	109,148
					Cooking	25,982	16,053	59,158
					Cleaning	10,988	4,381	24,112
RO	19,818	4,333	39,795	899	Balcony	8,974	4,333	11,627
					Garden	21,564	9,625	39,795
OP	32,415	27,979	36,853	4,076	Other	32,415	27,979	36,853

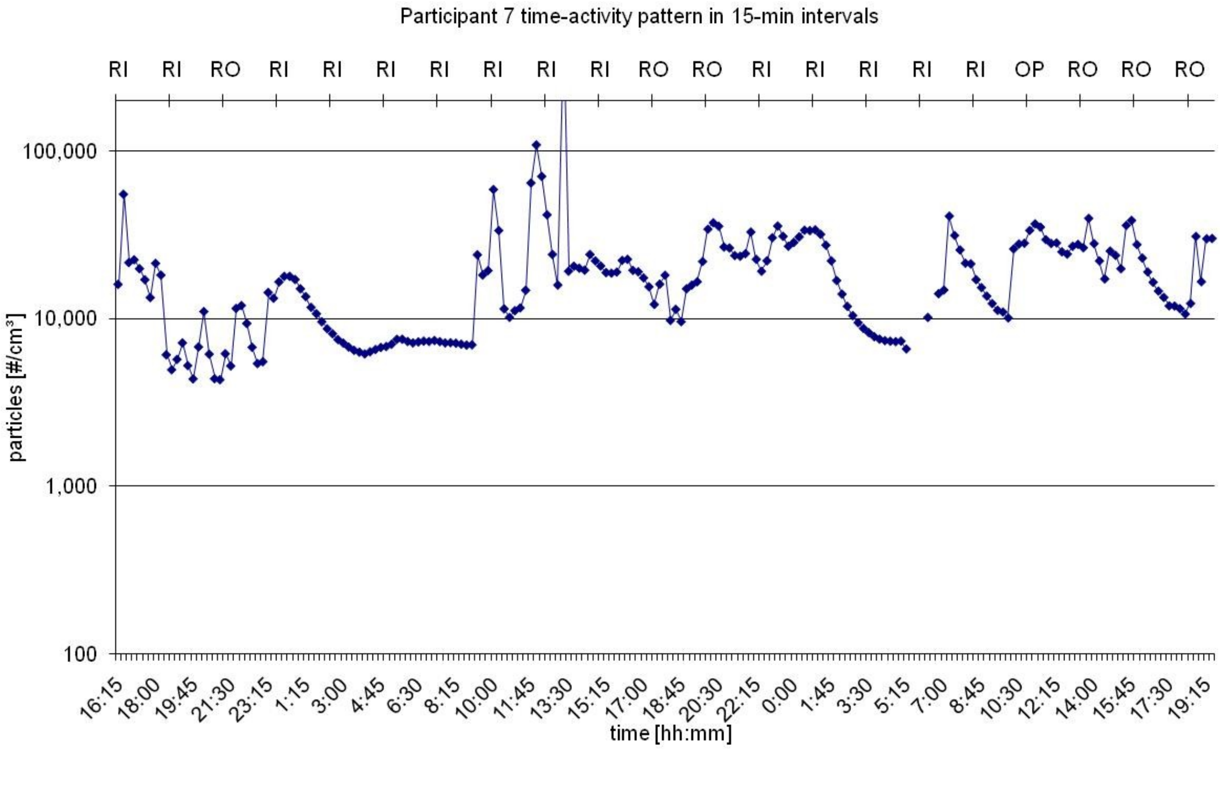

Fig. III.7 Participant 7 time-activity pattern

Participant 8

Tab. III.15 time-pattern participant 8

Age: 47	Sex: male		
ME	Time [h]	Activity/location	Time [h]
IT	4.5[1]	Car	4.5
RI	19.5[2]	Home	18.25
		Ventilating	1.25
AW	20[3]	Office	20
OP	1[4]	Other	1

[1]measured 1h
[2]measured 18.25h
[3]measured 19.5h
[4]measured 0h

Tab. III.16 activity-pattern participant 8

ME	Particle concentration [#/cm³]				Activity/location	Particle concentration [#/cm³]		
	Mean	Min	Max	SD		Mean	Min	Max
IT	8,503	4,564	12,442	5,571	Car	8,503	4,564	5,571
RI	822	298	2933	434	Home	838	333	2,933
					Ventilating	616	289	1,100
AW	7,255	2,893	15,362	2,680	Office	7,255	2,893	15,362

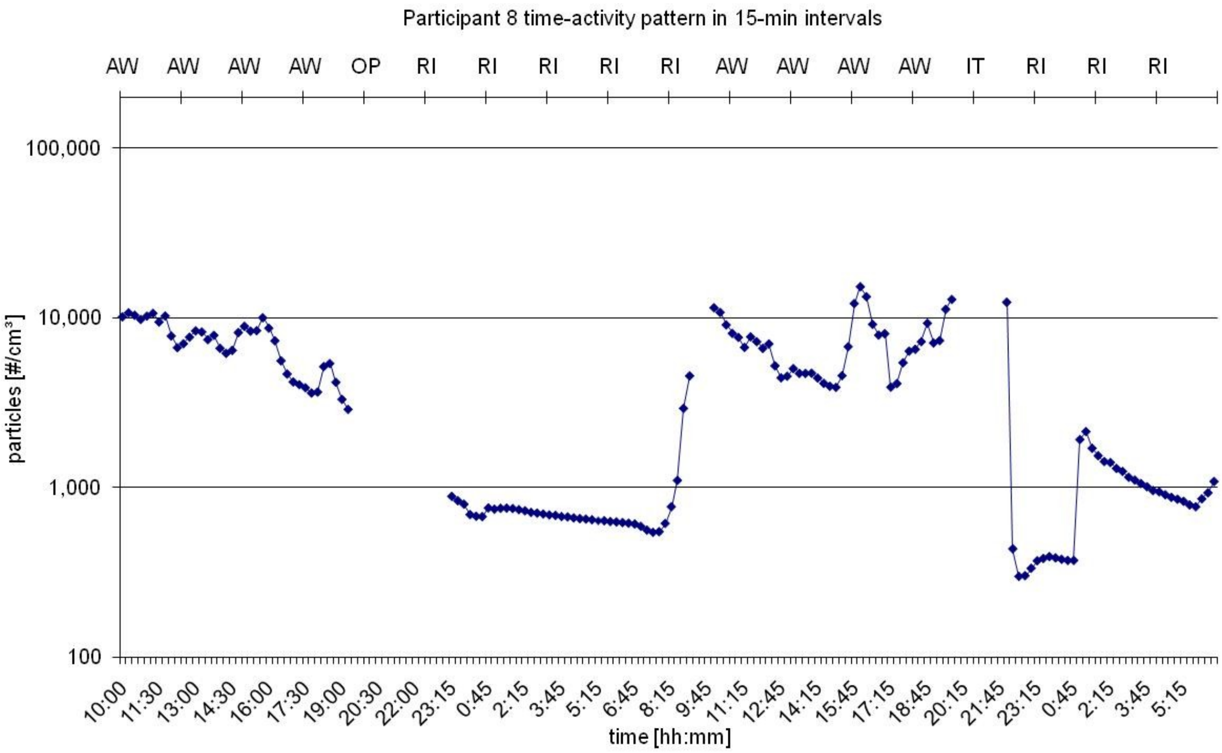

Fig. III.8 Participant 8 time-activity pattern

Participant 9

Tab. III.17 time-pattern participant 9

Age: 44	Sex: female		
ME	Time [h]	Activity/location	Time [h]
IT	17.5	Car	17.25
		Walking	0.25
RI	21	Sleeping	20.75[1]
		Cooking	0.25
AW	17.25[2]	Car	17.25[2]
OP	5.5	Other	5.5

[1] total time while sleeping 20.75 h, recorded 7.75 h
[2] AW and IT microenvironments have same values, thus participant spends more than 80% of the working time in traffic

Tab. III.18 activity-pattern participant 9

ME	Particle concentration [#/cm³]				Activity/location	Particle concentration [#/cm³]		
	Mean	Min	Max	SD		Mean	Min	Max
IT	2,267	573	9,331	1,810	Car	2,165	537	7,689
					Walking	9,331	9,331	9,331
RI	1,765	597	8,627	1,432	Sleeping	992	770	1,321
					Home	1,894	597	8,267
					Cooking	1,018	1,018	1,018
OP	6,889	1,862	16,331	4,654	Other	6,889	1,862	16,331

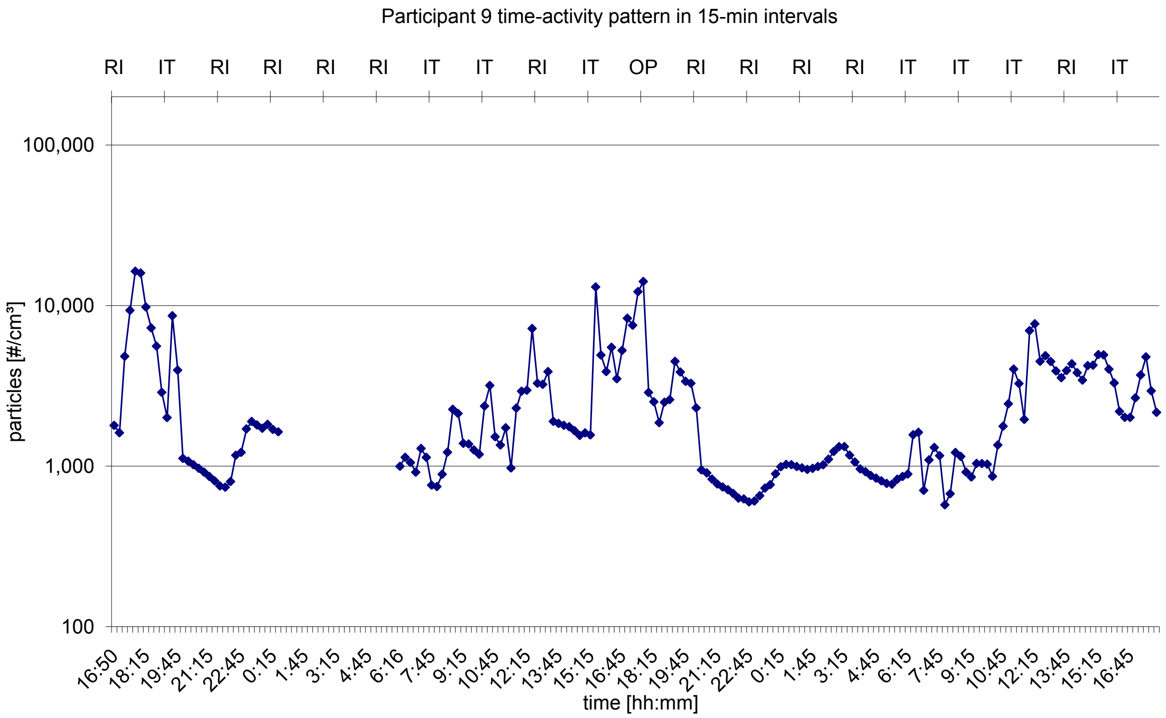

Fig. III.9 Participant 9 time-activity pattern

Participant 10

Tab. III.19 time-pattern participant 10

Age: 35	Sex: female		
ME	Time [h]	Activity/location	Time [h]
IT	3.25	Car	3.25
RI	25.75	Sleeping	17
		Ventilating	0.75
		Home	7.5
		Cleaning	0.5
AW	17.25	Office	17.25

Tab. III.20 activity-pattern participant 10

ME	Particle concentration [#/cm³]				Activity/location	Particle concentration [#/cm³]		
	Mean	Min	Max	SD		Mean	Min	Max
IT	16,639	2,570	19,194	3,200	Car	16,639	2,570	19,194
RI	5,355	2,170	33,715	3,672	Sleeping	4,550	2,600	8,339
					Home	5,599	2,170	15,115
					Cleaning	26,477	19,240	33,715
					Ventilating	7,085	5,250	8,285
AW	6,228	2,570	19,195	3,200	Office	6,228	2,570	19,195

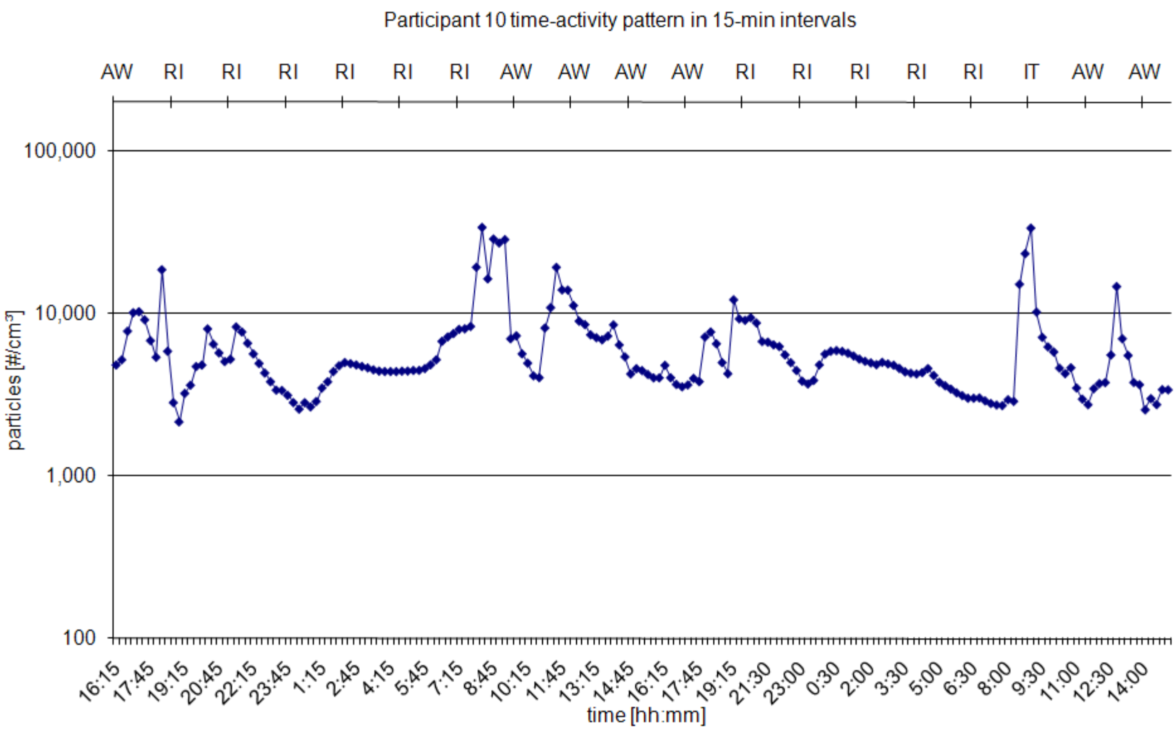

Fig. III.10 Participant 10 time-activity pattern

Participant 11

Tab. III.21 time-pattern participant 11

Age: 69	Sex: male		
ME	Time [h]	Activity/location	Time [h]
IT	4.25	Car	2.25
		Bicycle	0.75
		Walking	1.25
RI	32	Sleeping	20.25
		Candles	1
		Ventilating	0.5
		Cooking/ventilating	1,5
		Home	8.75
RO	9.75	Garden	7
		Garden/woodburning	2.75

Tab. III.22 activity-pattern participant 11

ME	Particle concentration [#/cm³]				Activity/location	Particle concentration [#/cm³]		
	Mean	Min	Max	SD		Mean	Min	Max
IT	21,608	4,418	11,1573	25,680	Car	29,400	8,041	111,573
					Walking	9,462	4,418	19,507
					Bicycle	18,476	10,017	29,783
RI	15,546	3,810	82,118	17,486	Sleeping	8,038	3,810	45,042
					Home	23,367	4,955	82,118
					Candles	10,294	7,917	12,553
					Ventilating	9,074	8,858	9,291
					Cooking/ventilating	33,065	6,251	59,675
RO	17,137	17,137	4,157	68,213	Garden	13,208	4,157	68,213
					Garden/woodburning	27,137	6,529	33,579

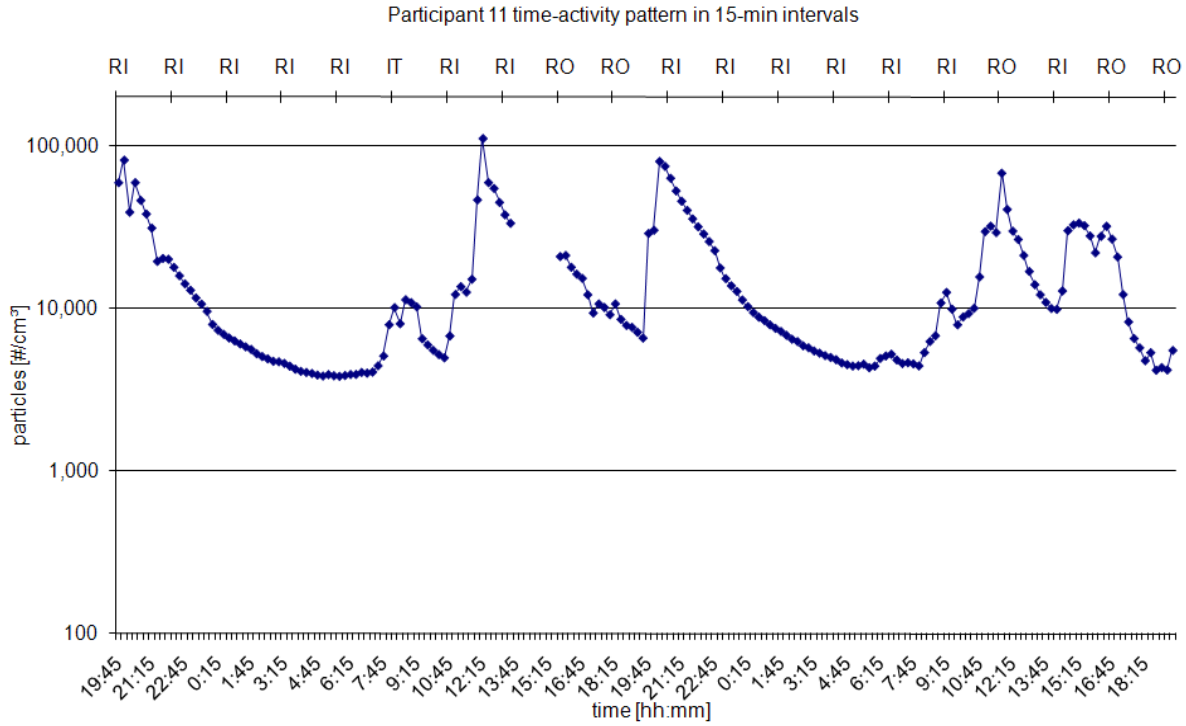

Fig. III.11 Participant 11 time-activity pattern

Participant 12

Tab. III.23 time-pattern participant 12

Age: 68 ME	Sex: female Time [h]	Activity/location	Time [h]
IT	1	Walking	1
RI	39.75	Sleeping	20
		Home	12.25
		Ventilating	1
		Cooking	1
		Cooking/ventilating	3.5
		Cleaning/ventilating	2
RO	7.5	Garden/woodburning	4.5
		Garden	3

Tab. III.24 activity-pattern participant 12

ME	Particle concentration [#/cm³]				Activity/location	Particle concentration [#/cm³]		
	Mean	Min	Max	SD		Mean	Min	Max
IT	7,259	5,517	10,829	2,502	Walking	7,259	5,517	10,829
RI	8,673	2,374	99,041	11,305	Sleeping	4,536	2,374	15,840
					Home	10,165	3,251	42,633
					Cooking	30,222	4,925	99,041
					Cleaning	7,311	3,221	21,151
					Cleaning/ventilating	5,338	4,898	6,246
					Cooking/ventilating	21,959	4,830	76,848
RO	7,292	3,341	48,799	8,098	Garden	9,119	3,341	48,799
					Garden/woodburning	6,073	1,593	11,818

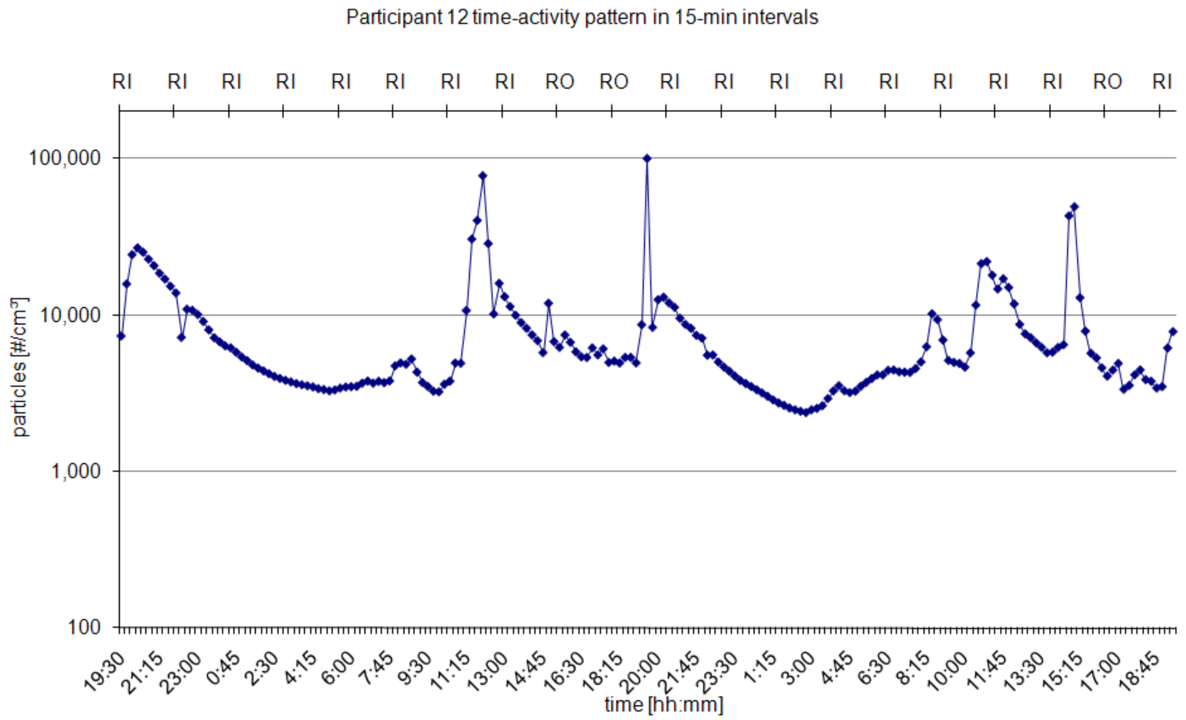

Fig. III.12 Participant 12 time-activity pattern

Participant 13

Tab. III.25 time-pattern participant 13

Age: 50	Sex: female		
ME	Time [h]	Activity/location	Time [h]
IT	9.75	Car	7.5
		Walking	2.25
RI	39	Sleeping	13.25
		Cleaning	3.5
		Ventilating	1.75
		Cooking	1.25
		Candles	0.5
		Home	18.75
AW	1.25	Office	1.25
RO	2	Garden	2

Tab. III.26 activity-pattern participant 13

ME	Particle concentration [#/cm³]				Activity/location	Particle concentration [#/cm³]		
	Mean	Min	Max	SD		Mean	Min	Max
IT	14,464	2,594	50,159	12,850	Car	16,021	2,594	50,159
					Walking	9,273	6,191	18,374
RI	9,547	3,086	112,161	15,064	Sleeping	5,317	3,425	11,665
					Home	14,980	3,133	121,161
					Cooking	62,666	6,524	94,158
					Cleaning	15,050	3,086	75,768
					Ventilating	5,876	5,125	6,903
					Candles	14,504	12,749	16,259
RO	18,016	5387	75,768	23,831	Garden	18,016	5,387	75,768
AW	4,182	3,722	4,850	494	Office	4,182	3,722	4,850

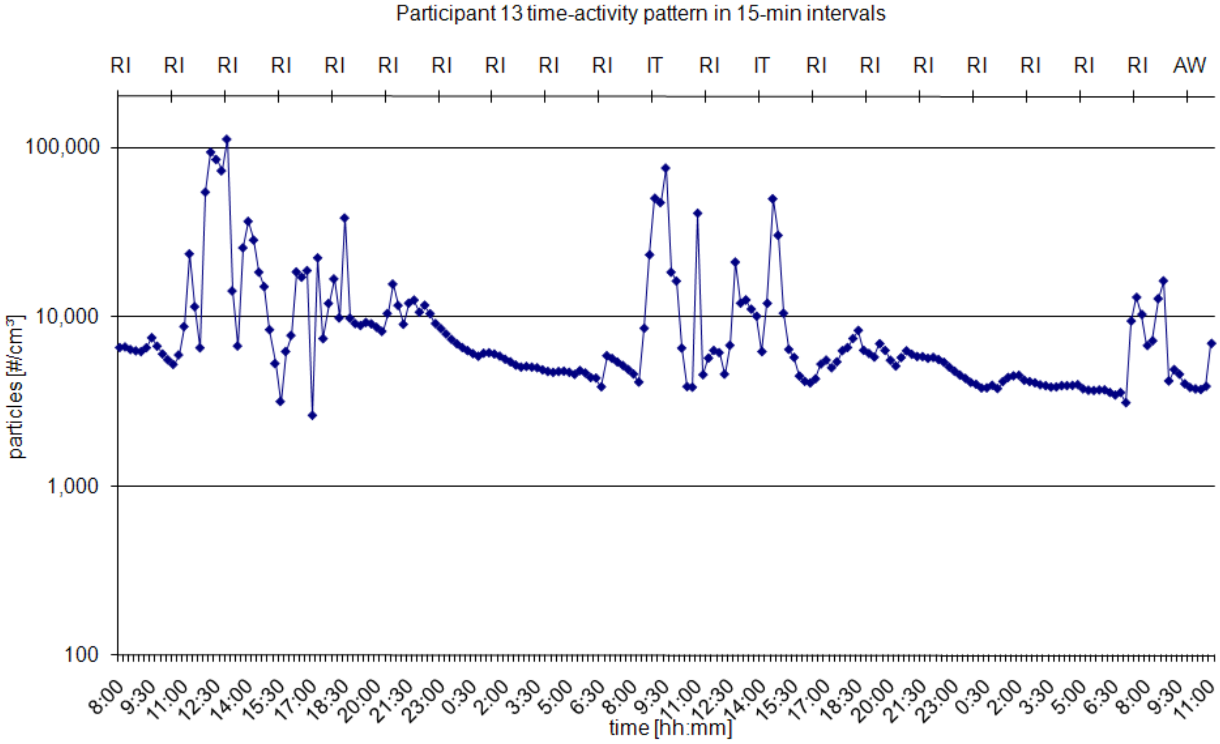

Fig. III.13 Participant 13 time-activity pattern

Participant 14

Tab. III.27 time-pattern participant 14

Age: 43	Sex: female		
ME	Time [h]	Activity/location	Time [h]
IT	0.75	Bicycle	0.75
RI	55.75	Sleeping	15
		Ventilating	12.75
		Cooking	0.75
		Cooking/ventilating	1.25
		Home	24.5
		Cleaning	1.5
RO	2.25	Garden	2.25

Tab. III.28 activity-pattern participant 14

ME	Particle concentration [#/cm³]				Activity/location	Particle concentration [#/cm³]		
	Mean	Min	Max	SD		Mean	Min	Max
IT	10,820	10,543	11,225	358	Bicycle	10,820	10,543	11,225
RI	9,678	1,476	174,489	18,202	Sleeping	3,348	1,476	9,274
					Home	6,765	1,035	44,446
					Cooking	15,189	7,859	29,737
					Cleaning	15,350	5,379	11,003
					Ventilating	18,099	2,035	174,489
					Cooking/ventilating	20,105	8,367	59,600
RO	8,645	6,503	11,457	1,783	Garden	8,645	6,503	11,457

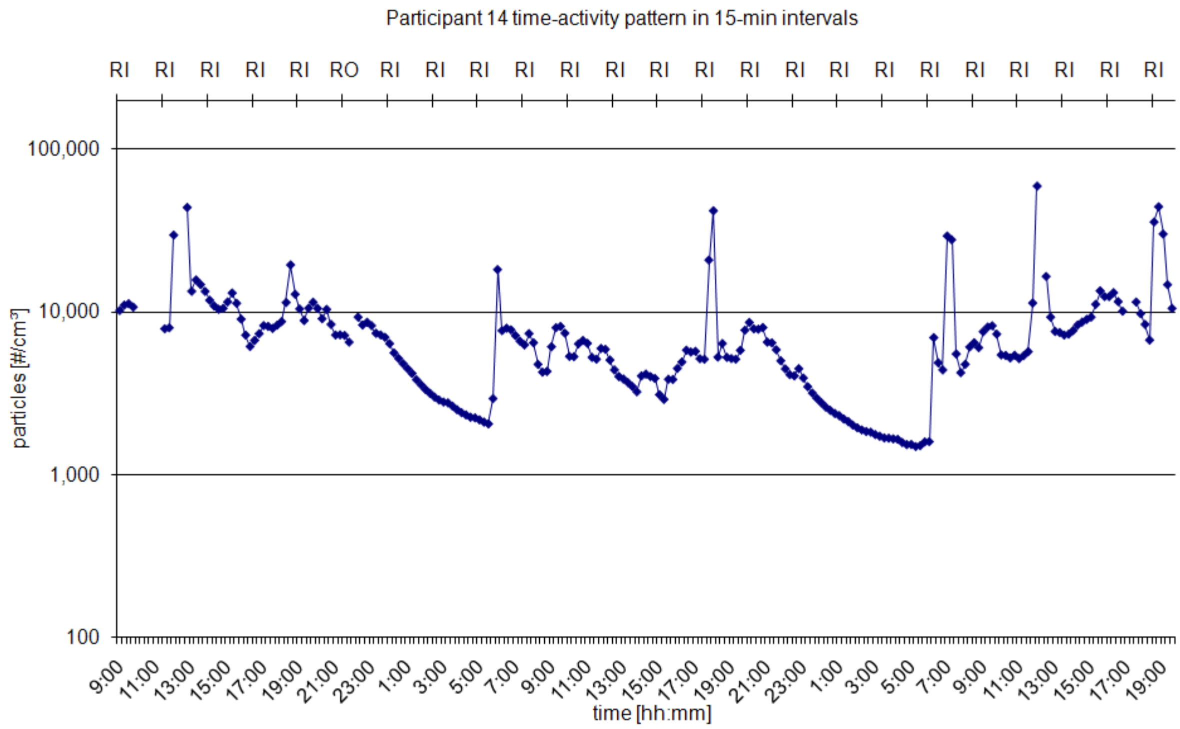

Fig. III.14 Participant 14 time-activity pattern

Participant 15

Tab. III.29 time-pattern participant 15

Age: 49	Sex: male		
ME	Time [h]	Activity/location	Time [h]
IT	1.25	Bicycle	1.25
RI	37	Sleeping	14.8
		Cleaning	3
		Cooking	0.5
		Home	18.8
AW	10	Office/traffic exposure	10

Tab. III.30 activity-pattern participant 15

ME	Particle concentration [#/cm³]				Activity/location	Particle concentration [#/cm³]		
	Mean	Min	Max	SD		Mean	Min	Max
IT	8,986	4,397	11,442	3,977	Bicycle	8,986	4,397	11,442
RI	13,187	2,843	12,745	17,004	Sleeping	7,616	2,843	83,906
					Home	14,835	7,423	112,726
					Cleaning	13,079	7,381	19,711
					Cooking	91,498	55,540	127,457
AW	13,729	4,733	26,043	6,319	Office/lorry	13,729	4,733	26,043

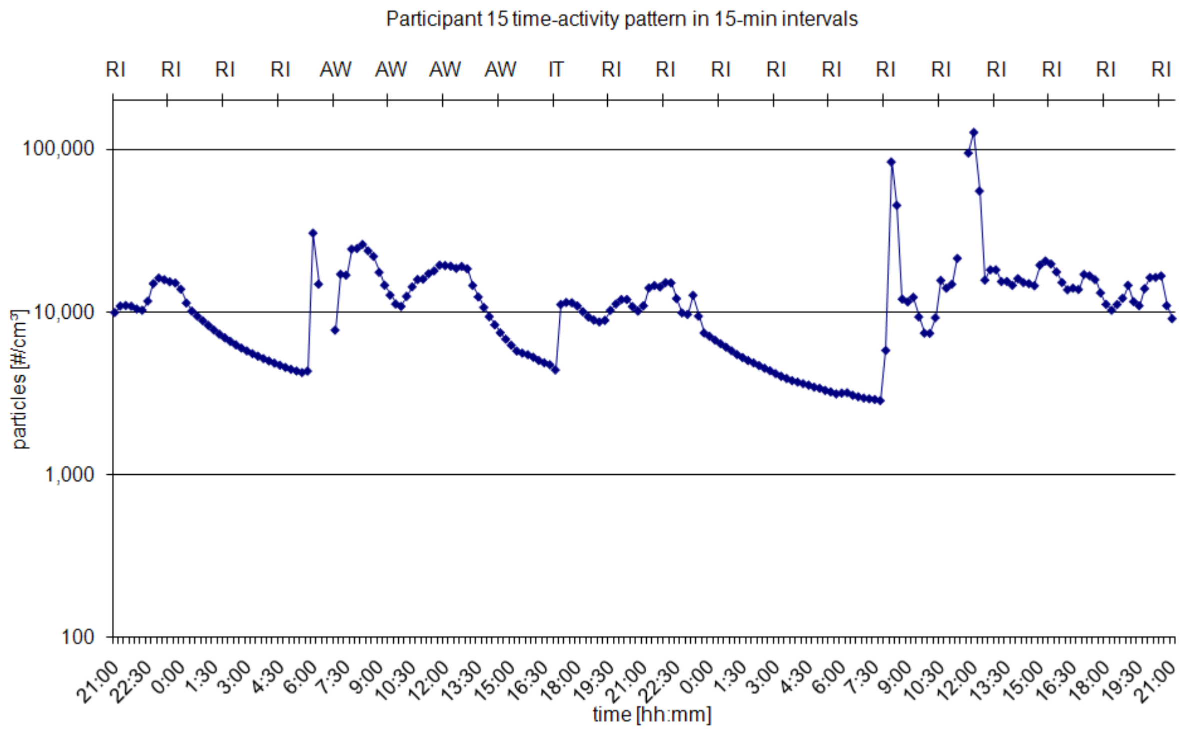

Fig. III.15 Participant 15 time-activity pattern

Participant 16

Tab. III.31 time-pattern participant 16

Age: 74	Sex: male		
ME	Time [h]	Activity/location	Time [h]
IT	0.75	Car	0.75
RI	43.75	Sleeping	18.5
		Home	19.5

Tab. III.32 activity-pattern participant 16

ME	Particle concentration [#/cm³]				Activity/location	Particle concentration [#/cm³]		
	Mean	Min	Max	SD		Mean	Min	Max
IT	15,661	8,539	26,197	9,311	Car	15,661	8,539	26,197
RI	9,985	3,898	54,834	6,873	Sleeping	9,297	3,898	14,762
					Home	10,983	4,136	54,843
					Cleaning	7,195	4,912	8,817
					Ventilating	9,506	4,644	17,687
					Cleaning/ventilating	6,797	5,613	8,254

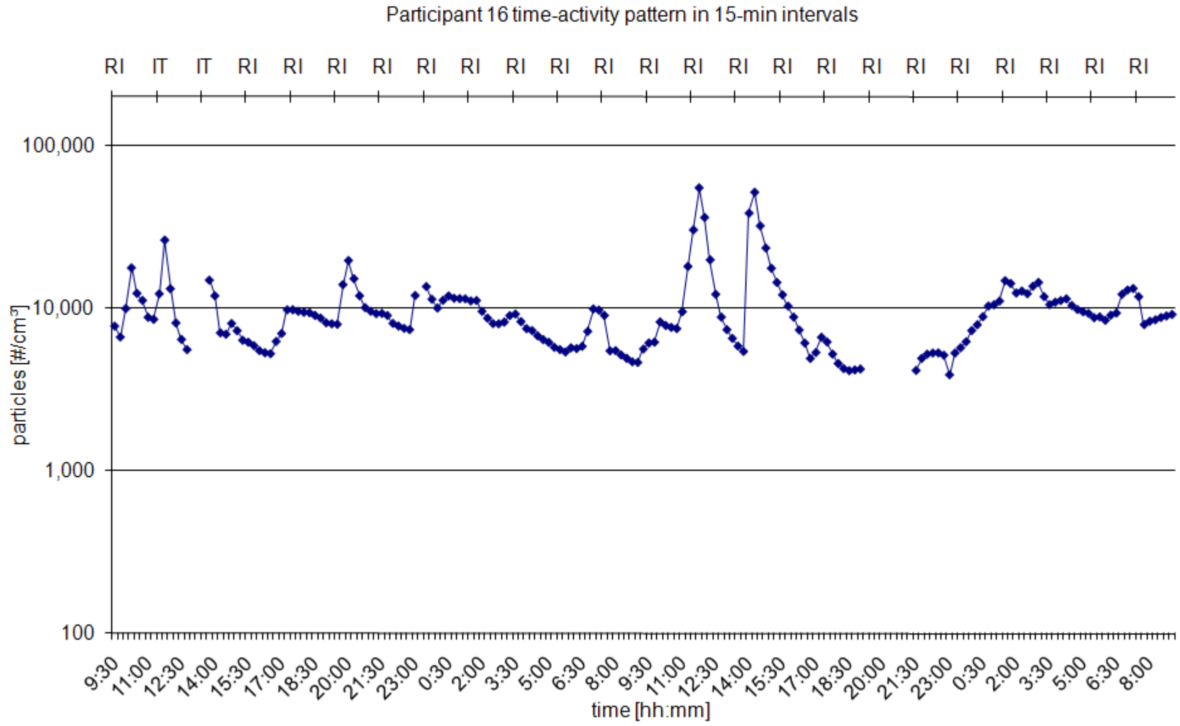

Fig. III.16 Participant 16 time-activity pattern

Participant 17

Tab. III.33 time-pattern participant 17

Age: 51	Sex: female		
ME	Time [h]	Activity/location	Time [h]
IT	1.75	Car	1.75
RI	43.75	Sleeping	18.0
		Home	19.0
		Cleaning	2,25
		Cooking	4.5
RO	3	Garden	3

Tab. III.34 activity-pattern participant 17

ME	Particle concentration [#/cm³]				Activity/location	Particle concentration [#/cm³]		
	Mean	Min	Max	SD		Mean	Min	Max
IT	6,500	2,875	20,991	7,655	Car	6,500	2,875	20,991
RI	11,033	5,880	9,495	1,230	Sleeping	6,178	2,837	16,059
					Home	10,326	2,758	17,894
					Cooking	12,846	3,843	80,000
					Cleaning	9,308	5,337	15,905
RO	7,726	2,348	14,036	3,777	Garden	7,726	2,348	14,036

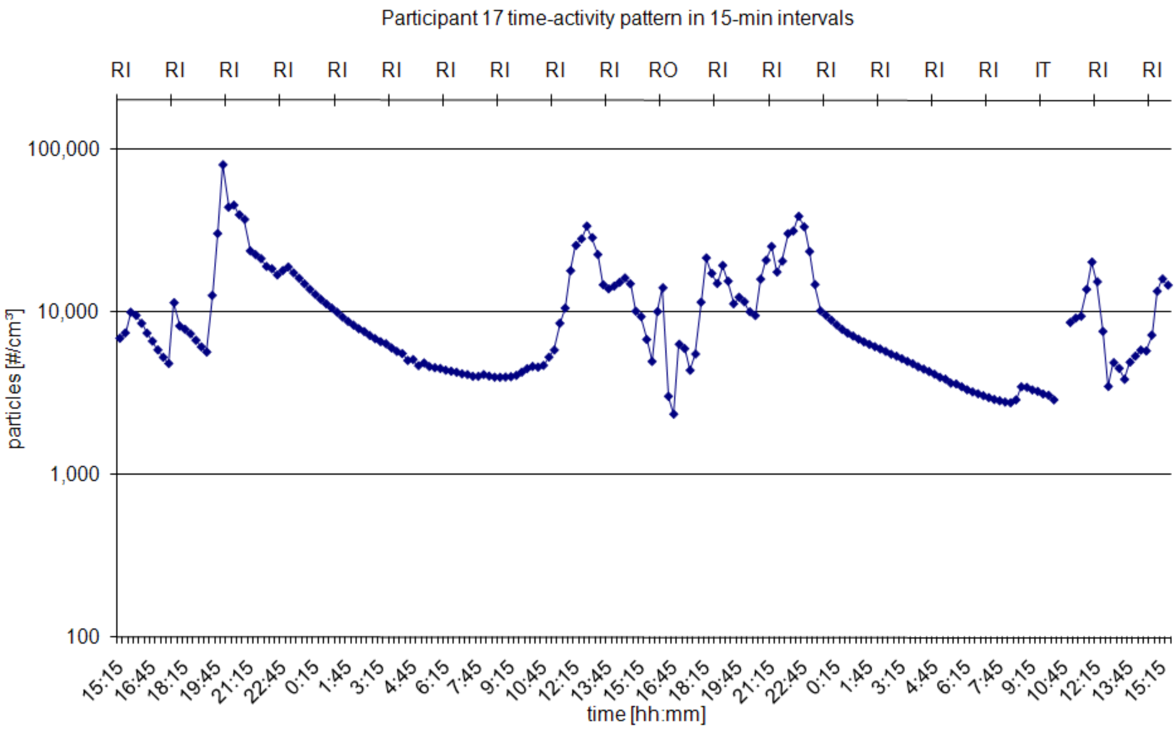

Fig. III.17 Participant 17 time-activity pattern

Participant 18

Tab. III.35 time-pattern participant 18

Age: 58	Sex: female		
ME	Time [h]	Activity/location	Time [h]
IT	2.25	Car	2.25
RI	31	Sleeping	15.5
		Home	14.5
		Cooking	1
AW	11.75	Surgery	11.75
RO	1	Garden	1
OP	2.75	Other	2.5
		Other/ETS	0.25

Tab. III.36 activity-pattern participant 18

ME	Particle concentration [#/cm³]				Activity/location	Particle concentration [#/cm³]		
	Mean	Min	Max	SD		Mean	Min	Max
IT	14,755	2,771	28,476	8,729	Car	14,755	2,771	28,476
RI	11,654	2,240	136,676	20,230	Sleeping	4,070	2,633	5,611
					Home	17,584	2,240	136,676
					Cooking	43,222	5,753	110,124
AW	14,873	7,614	33,911	6,680	Surgery	14,873	7,614	33,911
OP	26,344	17,901	53,465	10,665	Other	23,632	17,901	37,631
					Other/ETS	53,465	53,465	53,465
RO	9,998	9,064	12,282	1,530	Garden	9,998	9,064	12,282

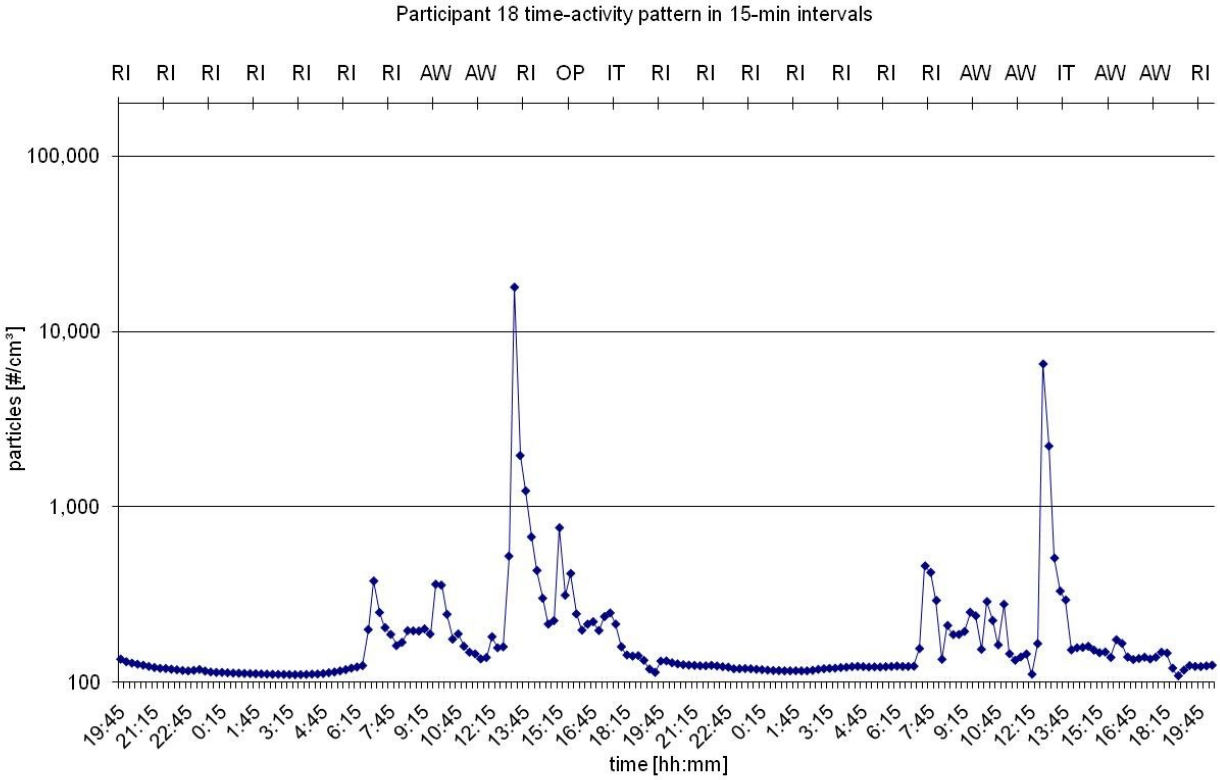

Fig. III.18 Participant 18 time-activity pattern

Participant 19

Tab. III.37 time-pattern participant 19

Age: 64	Sex: male		
ME		Activity/location	Time [h]
IT	5.5	Car	5.5
RI	35.25	Sleeping	16
		Home	13
		Cleaning	4.25
		Cooking	2
OP	7.5[1]	Other	7.5

[1]measured 6.75h

Tab. III.38 activity-pattern participant 19

ME	Particle concentration [#/cm³]				Activity/location	Particle concentration [#/cm³]		
	Mean	Min	Max	SD		Mean	Min	Max
IT	15,017	2,220	30,141	6,761	Car	15,017	2,220	30,141
RI	11,626	2,238	89,666	13,506	Sleeping	5,897	4,797	12,982
					Home	13,704	2,238	89,666
					Cooking	36,469	13,161	77,063
					Cleaning	15,151	8,022	39,384
OP	14,144	5,292	35,052	6,911	Other	14,144	5,292	35,052

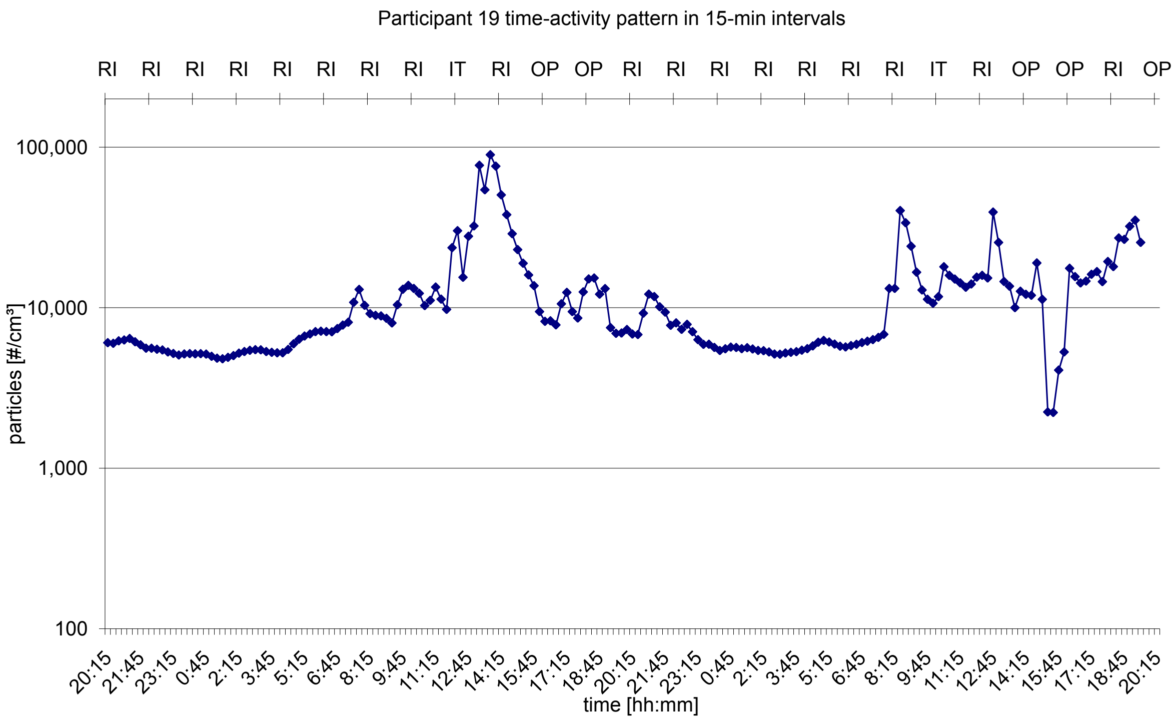

Fig. III.19 Participant 19 time-activity pattern

Participant 20

Tab. III.39 time-pattern participant 20

Age: 50	Sex: female		
ME	Time [h]	Activity/location	Time [h]
IT	4.5[1]	Bicycle	3.25[1]
		Walking	1.25
RI	25[2]	Home	24.75[3]
		Candles	0.25
AW	20.25	Office	17.5[4]
		Office/ETS	0.25
		Office/ventilating	2.5[5]
RO	0.25	Garden	0.25

[1]measured time 4.25h [2]measured time 21.5h [3]measured time 21.5h
[4]measured time 13.5h [5]measured time 1.75h

Tab. III.40 activity-pattern participant 20

ME	Particle concentration [#/cm³]				Activity/location	Particle concentration [#/cm³]		
	Mean	Min	Max	SD		Mean	Min	Max
IT	12,917	6,640	29,654	6,029	Bicycle	11,827	7,808	22,674
					Walking	15,533	6,640	29,654
RI	17,727	6,192	102,909	14,748	Candles	38,411	38,411	38,411
					Home	14,452	6,192	102,909
AW	10,471	5,489	26,303	4,864	Office	10,782	5,536	26,303
					Office/ETS	15,003	15,003	15,003
					Office/ventilating	7,419	5,489	10,972
RO	33,866	33,866	33,866	-	Garden	33,866	33,866	33,866

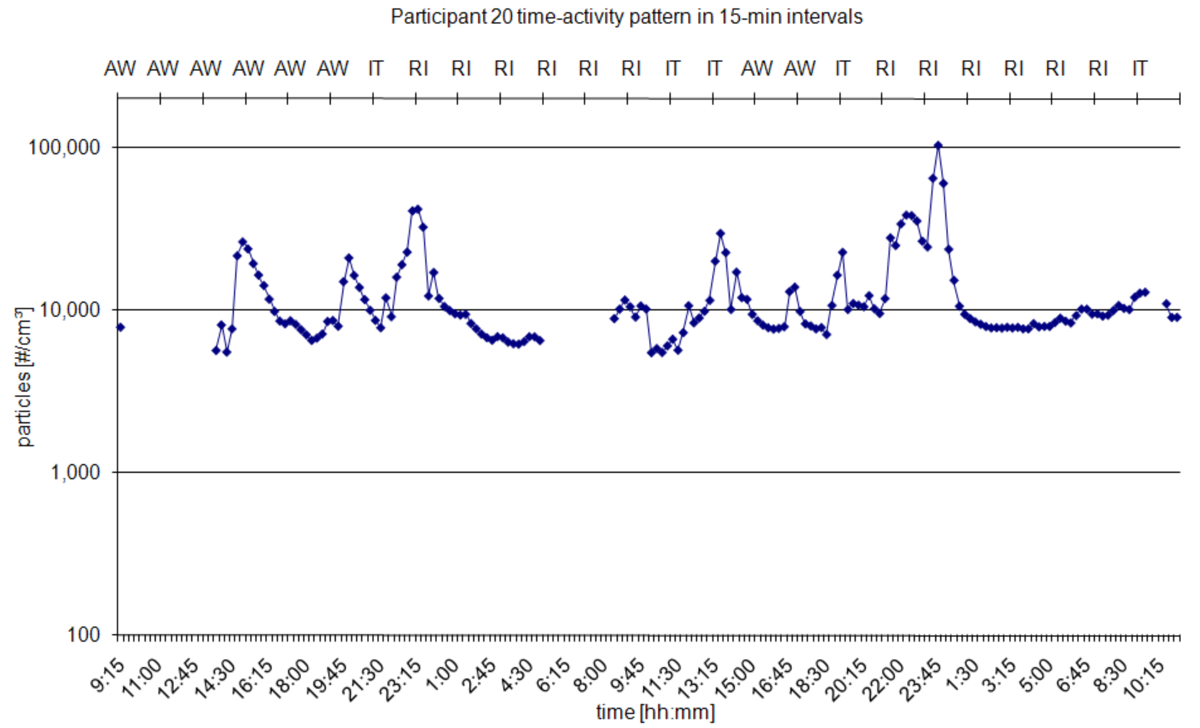

Fig. III.20 Participant 20 time-activity pattern

Participant 21

Tab. III.41 time-pattern participant 21

Age: 53	Sex: male		
ME	Time [h]	Activity/location	Time [h]
IT	2.25[1]	Car	1.25
		Bicycle	1
RI	33.5[2]	Home	33.5[2]
AW	9	Hospital	9
RO	3.25[3]	Garden	3.25[3]

[1]measured 2h [2]measured 29.25h [3]measured 1.5h

Tab. III.42 activity-pattern participant 21

ME	Particle concentration [#/cm³] Mean	Min	Max	SD	Activity/location	Particle concentration [#/cm³] Mean	Min	Max
IT	12,396	5,237	23,765	5,698	Car	15,349	10,783	23,765
					Bicycle	7,474	5,237	10,268
RI	7,877	3,235	4,289	27,584	Home	7,877	3,235	4,289
RO	6,379	5,772	6,794	423	Garden	6,379	5,772	6,794
AW	6,838	4,613	11,942	2,121	Hospital	6,838	4,613	11,942

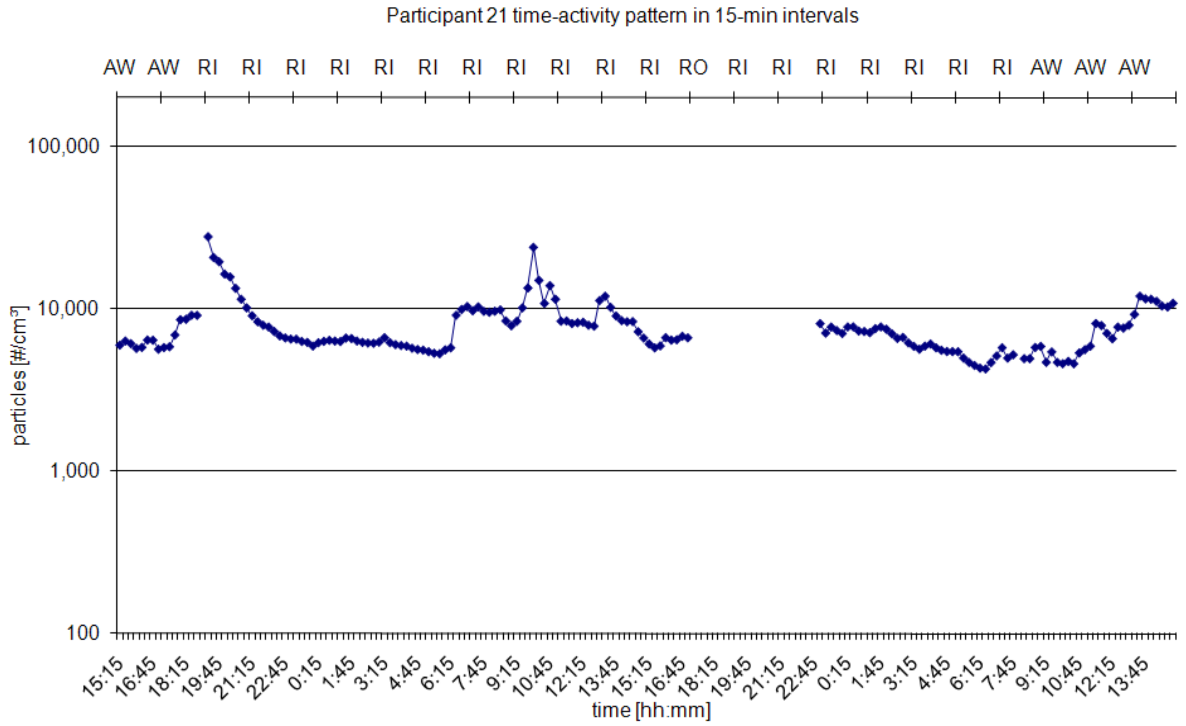

Fig. III.21 Participant 21 time-activity pattern

Participant 22

Tab. III.43 time-pattern participant 22

Age: 52 ME	Sex: female Time [h]	Activity/location	Time [h]
IT	1.75	Car	1.25
		Walking	0.5
RI	36.75[1]	Sleeping	8³
		Cooking	1[5]
		Cooking/ventilating	0.5[6]
		Home	9[4]
		Ventilating	17.75
RO	8.25²	Garden	8.25²
OP	2.75	Other	2.75

[1]measured 14.5h ²measured 6.25h ³measured 0h
[4]meaured 5.5h [5]measured 0.5h [6]measured 0h

Tab. III.44 activity-pattern participant 22

ME	Particle concentration [#/cm³] Mean	Min	Max	SD	Activity/location	Particle concentration [#/cm³] Mean	Min	Max
IT	2,588	726	4,739	1,262	Car	2,648	725	4,739
					Walking	2,438	1,826	3,051
RI	6,186	1,337	29,571	3,573	Sleeping	-	-	-
					Home	6,306	1,337	5,665
					Cooking	4,806	4,645	4,966
					Ventilating	6,467	4,398	9,166
					Cooking/ventilating	-	-	-
RO	3,813	2,690	5,574	751	Garden	3,813	2,690	5,574
OP	777	632	1,057	1,262	Other	777	632	1,057

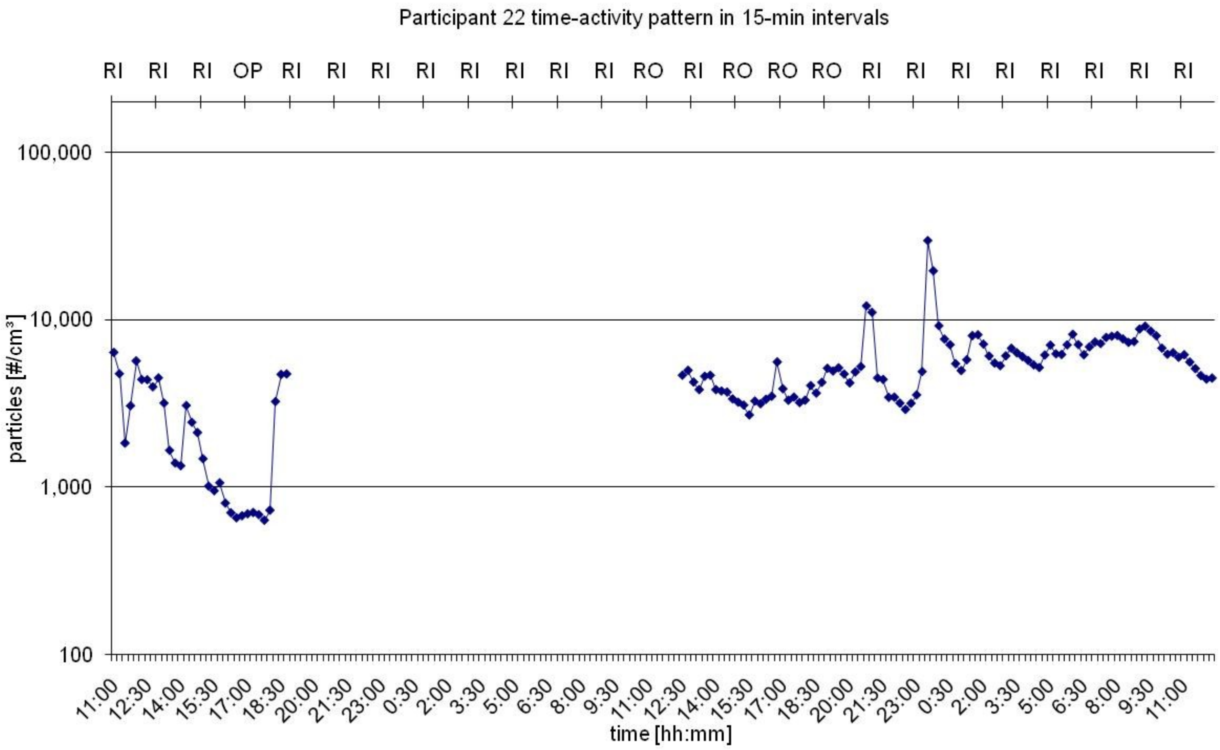

Fig. III.22 Participant 22 time-activity pattern

Curriculum Vitae

Der Lebenslauf ist in der Online-Version aus Gründen des Datenschutzes nicht enthalten.

Acknowledgements

First and foremost, I would like to thank **PROF. DR. K.-H. JÖCKEL** and **PD DR. B. HOFFMANN** for their enthusiastic supervision and support and for granting permission to carry out this thesis at the **UNIVERSITY HOSPITAL OF ESSEN (UNIVERSITY OF DUISBURG-ESSEN, DEPARTMENT OF MEDICAL INFORMATICS, BIOMETRICS AND EPIDEMIOLOGY)**. I am also grateful to the entire research team at the IMIBE

I would like to thank **PROF. DR. DR. H.C. R. ZELLNER (DEPARTMENT OF PHYSICS AND THEORETICAL CHEMISTRY, UNIVERSITY OF DUISBURG-ESSEN)** whose professional expertise was invaluable during this research project.

Special thanks go to **DR. T. KUHLBUSCH** (Institute of Energy and Environmental Technology) and his staff. I am grateful to **DR. O. HÄNNINEN** for helping me prepare the experiments and offering valuable advice whenever needed, and to **MS. A. MARR** for her help with SAS programming.

I would also like to extend my thanks to **DR. M. PISANI**, **DR. M. ZIMMERMANN** and **DR. T. FARRUGIA** for proofreading this thesis.

I am indebted to all the volunteers who participated in this study without whose help this research would not have been possible.

Sincere and heartfelt thanks go to my family, especially to my parents and brother, as well as to my friends for their understanding, forbearance and endless encouragement. Finally, I would also like to thank all those individuals who, in one way or another, offered me help and support.

Erklärung:

Hiermit erkläre ich, gem. § 6 Abs. 2, Nr. 7 der Promotionsordnung der Math.-Nat. Fakultäten zur Erlangung des Dr. rer. nat., dass ich das Arbeitsgebiet, dem das Thema *„Pilot Study for the Development of a Personal Exposure Model for Personal Ultrafine Particle Exposure“* zuzuordnen ist, in Forschung und Lehre vertrete und den Antrag von *Frank Pisani* befürworte.

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