Exposure of an Urban Adult Population to $PM_{2.5}$

Methods, Determinants and Sources

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ACADEMIC DISSERTATION

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ABSTRACT

This thesis is based on data collected as a component of the population based *EXPOLIS* study in Helsinki metropolitan area during 1996-1997. The aims of the study were to assess precision of the applied monitoring methods, to quantify personal exposures of the adult urban population to $PM_{2.5}$ and to evaluate the applicability of ambient monitoring methods in assessing personal exposures to $PM_{2.5}$. Personal exposures were estimated directly with personal monitoring of 201 randomly selected study participants for 48-hour weekday sampling periods, and indirectly by measuring residential indoor, residential outdoor, and workplace $PM_{2.5}$ concentrations. In order to quantify contributions of each microenvironment to personal exposures, residential and workplace concentrations were monitored only during the times that the participants themselves reported that they would be present in the microenvironment during the 48-hour sampling period. Determinants of exposures were analysed using statistical methods. Subsequently, elemental analysis by ED-XRF was performed on personal exposure and microenvironment samples to identify and quantify $PM_{2.5}$ mass sources using principal component analysis and mass reconstruction techniques.

The precision of personal (PEM) and microenvironment (MEM) $PM_{2.5}$ monitors used in this study was equal or better than those in previous exposure studies. The most important sources of correctable error in gravimetric analysis of the blank filters were air buoyancy variation and filter storage time.

Personal 48-hour exposures to $PM_{2.5}$ in Helsinki were low compared to those that have been measured in most studies in North America and Europe. The geometric mean of cross sectional 48-hour population exposure across individuals and time was 10.9 $\mu g/m^3$ with geometric standard deviation of 2.13 $\mu g/m^3$. This population exposure level did not exceed the $PM_{2.5}$ National Ambient Air Quality Standards given in 1997 in the USA.

Presence of smoking was the most important determinant of $PM_{2.5}$ personal exposure concentrations and smoking, occupational status, education and age were the best predictors of differences in $PM_{2.5}$ exposures between socio-demographic sub-populations. Henceforward, all conclusions concern individuals who did not smoke and reported no exposure to environmental tobacco smoke (ETS). For participants not exposed to ETS residential outdoor and ambient fixed site $PM_{2.5}$ concentrations did not adequately predict exposures. Residential and workplace indoor concentrations combined with the traffic density on the streets next to residences were the best behavioural and environmental determinants of personal exposure concentrations.

The major source categories of $PM_{2.5}$ exposures identified in this study were smoking, inorganic soil components, primary and secondary long-range transported particles, and local combustion, mostly from traffic. For participants not exposed to ETS soil dust was the major indoor source and therefore ambient monitoring under-estimated exposures to soil particles. In contrast, use of ambient monitoring to predict personal exposures would over-estimate the contribution of particles from outdoor sources, and personal exposures to inorganic secondary particles, for example, would be overestimated by approximately two thirds.

Ambient total $PM_{2.5}$ mass variation was a moderate predictor of $PM_{2.5}$ exposure variation within the population (regression $r^2 = 0.4$). When total $PM_{2.5}$ mass was apportioned into contributions from each of the main sources, ambient monitoring of inorganic secondary PM was a good ($r^2 = 0.8$) predictor of inorganic secondary PM exposures. Similarly ambient monitoring of CoPM was a moderate predictor ($r^2 = 0.3$), and ambient monitoring of soil PM a weak predictor ($r^2 = 0.2$) of respective exposures. As expected, total ambient $PM_{2.5}$ mass was generally a weaker predictor for exposure to PM from each of these 3 sources.

To Kaija, Sanna, Kai and Suvi

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During the course of this study I have also had some contact with life outside of science. I think I am now more experienced, among other things, in understanding how to survive in Koivukylä, how to buy profitable pizza coupons, how to declare a car for customs purposes and how important it is to play badminton. I wish to express my thanks to all my friends and relatives, for sharing their time with me and giving me chance not to think about work.

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Kimmo Koistinen

ABBREVIATIONS

BS	Black smoke
EE	Exposure efficiency. The fraction of released material leading to exposure.
ETS	Environmental tobacco smoke
CoPM	Combustion and other particulate matter (CoPM) defined as “Other particles” in mass reconstruction analysis including primary combustion particles, non-volatile organic particles, particles from tyre wear, etc.
LOD	Level of detection
LRT	Long-range transported particles
MEM	Microenvironment monitor
PCA	Principal component analysis
PEM	Personal monitor
PM	Particulate matter in air
$PM_{2.5}$	Particulate matter in air with a 50% cut-off aerodynamic diameter of 2.5 µm
PTEAM	The U.S. EPA Particulate Total Exposure Assessment Methodology Study
QA/QC	Quality assurance / Quality control
RSP	Respirable suspended particles
SR	Source reconstruction. In SR elemental concentrations of PM were combined with source fingerprint or trace element data to estimate the total mass of the PM from the analysed source.
XRF	X-ray fluorescence

LIST OF ORIGINAL PUBLICATIONS

This thesis is based on the following original publications, referred to in the text by the Roman numerals (I-V). Some previously unpublished results are also presented in this thesis.

I Koistinen KJ, Kousa A, Tenhola V, Hänninen O, Jantunen M, Oglesby L, Künzli N, Georgoulis L. Fine particle ($PM_{2.5}$) measurement methodology, quality assurance procedures and pilot results of the *EXPOLIS* study. *J Air Waste Manage Assoc* 1999;49:1212-20.

II Hänninen OO, Koistinen KJ, Kousa A, Keski-Karhu J, Jantunen MJ. Quantitative analysis of environmental factors in differential weighing of blank Teflon filters. *J Air Waste Manage Assoc* 2002;52:134-139.

III Koistinen KJ, Hänninen O, Rotko T, Edwards R, Moschandreas D, Jantunen MJ. Behavioral and environmental determinants of personal exposures to $PM_{2.5}$ in *EXPOLIS* -Helsinki, Finland. *Atmos Environ* 2001;35:2473-81.

IV Rotko T, Koistinen K, Hänninen O, Jantunen M. Sociodemographic descriptors of personal exposure to fine particles ($PM_{2.5}$) in *EXPOLIS*-Helsinki. *J Expo Anal Environ Epidemiol* 2000;10:385-93.

V Koistinen KJ, Edwards RD, Mathys P, Ruuskanen J, Künzli N, Jantunen MJ. Sources of fine particulate matter in personal exposures and residential indoor, residential outdoor and workplace microenvironments in the Helsinki phase of the *EXPOLIS* study. *Scand J Work Environ Health*, in press.

CONTENTS

1 INTRODUCTION

Polluted air has been associated with harmful health effects since the ninth century, when coal collected from the Northeast coast of England was burned as fuel (Wilson 1996). Edward I (1272-1307) banned the use of “sea coal” as a fuel in London and ordered people to replace sea coal with brushwood or charcoal when heating houses. Somewhat later, air pollution episodes drew public attention because of their serious health impacts in populations. Well-publicised episodes occurred first in Meuse Valley of Belgium in December 1930 and in London in December 1952. In both of these episodes mortality and morbidity increased significantly during the days of high pollution. After the London smog episode the availability of cleaner fuels, oil and natural gas, together with construction of higher chimneys that sent pollutants above the inversion layer, dramatically improved air quality in cities. By the 1980’s the local problem had been reduced in its prominence, but increasing concern was being voiced over global and trans-boundary issues including acid rain and global warming. Fine particles that had a high potential to be inhaled were being transported over large urban and rural areas as their small particle size resulted in long residence times suspended in the atmosphere. In the 1990's the adverse health effects that had been observed with airborne particulate matter (PM) with an aerodynamic diameter less than 10 µm were increasingly associated with the fine fraction of particles with an aerodynamic diameter less than 2.5 µm (Dockery et al. 1993, Pope et al. 1995, Schwarz et al. 1996, Pope et al. 1999, Schwarz and Neas 2000). The results of the Six-Cities-Study (Dockery et al. 1993, HEI 1995) and the APHEA study (Katsouyanni et al. 1997) have shown similar results in both USA and in Europe suggesting that health risks are a global concern. WHO estimated in 1995 that respirable and fine particulate matter (i.e. Particulate matter with 50% cut-off diameter at 10 µm (PM_{10}) and 2.5 µm ($PM_{2.5}$), respectively) may be responsible for tens of thousands of cases of respiratory and cardiovascular mortality annually and may significantly shorten life expectancies in Europe (WHO 1995).

Urban exposures to air pollutants have been assessed by ambient fixed site monitoring for decades. In addition to ambient air quality, researchers started to discuss the roles of behavioural (personal) and indoor sources in human exposure, which was defined as an event when a person comes to a contact with a pollutant (Ott 1982). This discussion culminated in probability based studies for total air pollution exposure assessment, otherwise known as the TEAM studies (CO –TEAM Akland et al. 1985, VOC-TEAM Wallace et al. 1988, PTEAM

Thomas et al. 1993 and Clayton et al. 1993) and the National Human Exposure Assessment Survey (NHEXAS), the first large-scale total (multi-route) exposure study in the USA (Lebowitz et al. 1995, Pellizzari et al. 1995). Subsequently, the largest American exposure study to evaluate exposures to $PM_{2.5}$ was carried out in Toronto, Canada during 1995-1996 (Pellizzari et al. 1999) and in Europe the first large-scale representative population exposure study for $PM_{2.5}$ was *EXPOLIS*, conducted in 1996 - 1998 (Jantunen et al. 1998), from which the current thesis is derived.

It is important that the adverse health effects in populations on a global basis as a result of exposure to $PM_{2.5}$ are linked to emissions in order to identify health impacts of particles from different sources. In order to focus administrative actions, exposures must be linked to specific sources and resultant health consequences, and both should be related to monetary costs to identify cost-effective strategies to reduce exposures. $PM_{2.5}$ exposure studies are crucial for obtaining this information. Currently there is not enough knowledge about determinants and predictors of exposure to $PM_{2.5}$ that allows us to relate exposures to their sources. It must be noted that the validity of ambient monitoring to predict exposure to $PM_{2.5}$ may be different for short-term or long-term effects of $PM_{2.5}$ or whether exposure to total $PM_{2.5}$ is concerned instead of exposure to source related particles.

This thesis is based on the studies carried out in Helsinki metropolitan area as a part of the *EXPOLIS* study and examines exposures to $PM_{2.5}$ in an urban adult population and evaluates the validity of ambient and indoor monitoring to assess source-related exposures to $PM_{2.5}$. The determinants identified in this thesis help modellers to choose valid input parameters when developing exposure models for $PM_{2.5}$. Recent studies (Laden et al. 2000, Pope et al. 2002) have indicated differences in health effects of fine particles of different composition. Thus, it is of current interest among the scientists to link sources to exposure and exposure of each source category to their health effects. The source apportionment results presented in this thesis link exposures of the main source categories of $PM_{2.5}$ to their sources and thus, offers a possibility to make the health risk assessment further by combining source-related exposure to health effects in forthcoming studies. Subsequently, these results can be used to set air quality guidelines, to assess the role of source categories to exposure, and to evaluate the validity of methods for the assessment of $PM_{2.5}$ exposures.

2 REVIEW OF THE LITERATURE

2.1 Exposure assessment

Human exposure assessment bridges the fields of traditional environmental science with toxicology and epidemiology (Lioy 1990). It searches for answers to three fundamental questions: 1) how one is exposed, 2) why one is exposed and 3) where and when one is exposed to a pollutant. Answers to these questions are necessary steps to reduce uncertainties in the link between emissions and health effects (Lioy 1999). Figure 1 shows the process continuum from emission of a contaminant to a health effect. Exposure assessment is thus an integral and a necessary part of health risk assessment.

Concentrations of pollutants may exist without ever coming into contact with humans. Adverse health effects of air pollutants can occur only if the contaminant has come to contact with an individual. Consequently contact is an essential component when defining the term 'exposure' and is defined, therefore, as "an event that occurs when a person comes into contact with a pollutant" (Ott 1982, Ott 1995). This definition differentiates exposure from concentration and dose. When a pollutant has crossed a physical boundary e.g. skin, or epithelial cell in mouth, nose or lung, the event has been defined as dose (Ott 1982). Exposure can occur without leading to dose, but dose is not possible without exposure.

The nature of exposure can be taken as an instantaneous event (Duan et 1989, Georgopoulos and Lioy 1994). In practise, however, exposure is measured over a certain time period, therefore mean or maximum exposures for this time period can be used to describe consecutive instantaneous exposures. Slightly different concepts of exposure have been defined in other publications (Duan 1982, NRC 1991, EPA 1992, Zartarian et al. 1997, Duan et al. 1989, Georgopoulos and Lioy 1994), and are reviewed comprehensively by Alm (1999) and Monn (2001). In this thesis "48-hour exposure to $PM_{2.5}$" has been used to refer to mean $PM_{2.5}$ exposure concentration for a 48-hour sampling period in units of concentration ($\mu g/m^3$) at the point of contact.

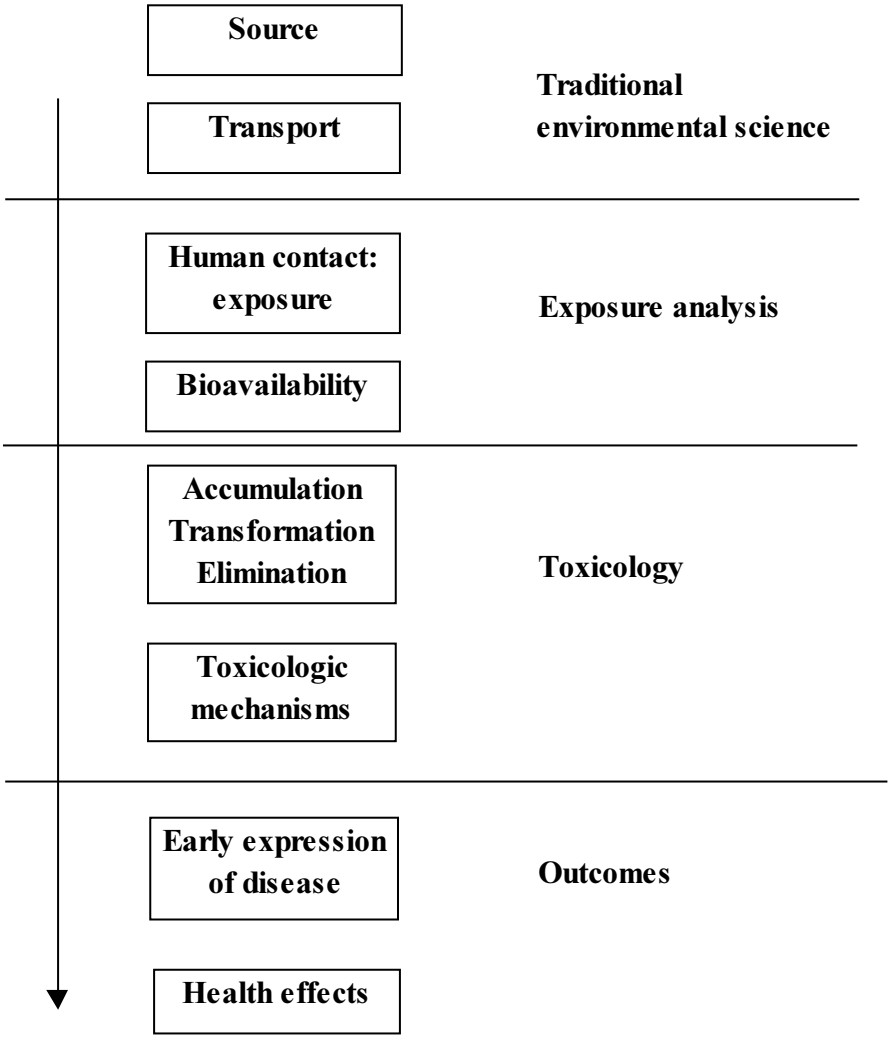

Figure 1. Frameworks of the exposure analysis (Adopted from Lioy 1990 and Lioy 1999)

The role of exposure assessment can be demonstrated using benzene in San Francisco, CA, USA, as an example (Ott 1995). In the early 1990's the Board of the Bay Area Air Quality Management District (BAAQMD) considered that benzene contributed the largest risk to inhabitants living in industrialised areas with numerous refineries and called for a 50% reduction in benzene emissions from industrial point sources. At the time the average outdoor benzene concentration was 2 $\mu g/m^3$. The TEAM study (Wallace et al. 1988), however, had previously demonstrated that, in the same area, average population exposures were far higher at 8 $\mu g/m^3$. Source apportionment of the benzene showed only 0.3 $\mu g/m^3$ of the total average population exposure was attributable to stationary sources. Thus, the reduction of 50% from stationary sources led to reduction of 0.15 $\mu g/m^3$ in average population exposure. Similar

reductions could have been achieved easily and cheaply for example by reducing exposure to benzene from passive smoking or spending time in traffic (Ott 1995).

Similarly, Smith (1993) reported that total emissions particles as environmental tobacco smoke (ETS) in the United States were equivalent to approximately 3% of the emissions from U.S. coal-fired power plants, but that the exposure relationship was totally different. A 1.3% reduction in exposure to ETS would be equivalent to elimination of the entire exposure to PM caused by coal-fired power plants. This example demonstrates how emissions from different sources reach human's breathing zones to different extents, resulting in different exposure efficiencies (EE) defined as fraction of released material leading to exposure (Smith 1993). Figure 2 shows the EEs for some source categories compared to the EE of coal-fired power plants for PM (adopted from Smith 1993). Thus, exposure of 1g of ETS was equivalent to human particulate exposure of 2400g released from a coal-fired power plant.

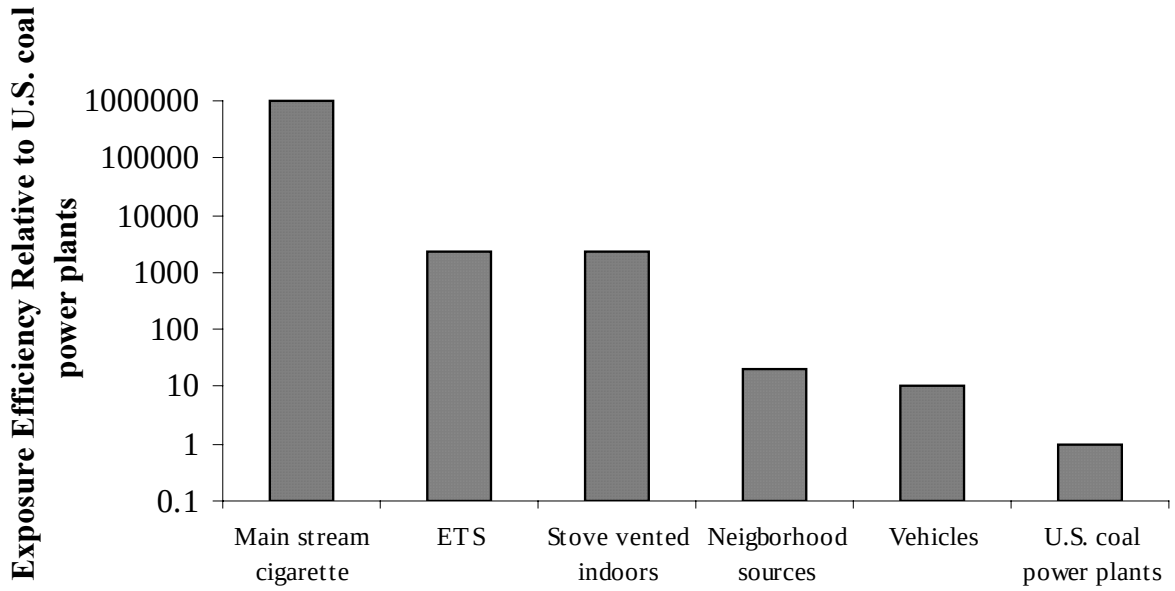

Figure 2. PM Exposure Efficiencies (EE) of different sources compared to the EE of the U.S. coal-fired power plants (adopted from Smith 1993).

2.2 Characteristics of $PM_{2.5}$

An aerosol is defined as a suspended mixture of solid or liquid particulate matter (PM) in surrounding gas containing many molecules held together by intermolecular forces and being primarily larger than molecular dimensions (Seinfeld and Pandis 1998). Aerodynamic diameter of a particle is defined as the diameter of a unit density (i.e. 1 g/cm^3) sphere that has the same settling velocity as the particle regardless of its shape, density or physical size. Aerodynamic diameter is a key parameter for characterising particle separation, filtration and respiratory deposition from air. (Hinds 1982). $PM_{2.5}$ refers to PM in which 50% of the particles have an aerodynamic diameter less than 2.5 µm.

$PM_{2.5}$ has been generally referred to as "fine particles" and those greater than $PM_{2.5}$ in diameter as "coarse particles" (Seinfeld and Pandis 1998). Fine and coarse particles differ from each other by origin, chemical composition and optical properties. In addition they are transformed separately, are removed from air by different mechanisms and finally have different deposition characteristics in the respiratory tract. Fine particles may stay in air from days to weeks, much longer than coarse particles, which settle down in minutes or hours. This also means that fine particles may travel up to 1000s of kilometres while coarse particles can usually only travel up to 10s of kilometres (Spengler and Wilson 1996). Thus, the nature of $PM_{2.5}$ is different from coarse particles and especially from gaseous air pollutants, which have the same physical, chemical and toxicological characteristics wherever they are present. These characteristics vary considerably between different samples of $PM_{2.5}$ and must be known before results from different $PM_{2.5}$ data sets can be compared.

2.3 Sources and determinants of $PM_{2.5}$ exposure

2.3.1 Emissions from ambient sources

Major components of fine particles in ambient air are: sulphate, strong acid, ammonium, nitrate, organic compounds, trace elements (including metals), elemental carbon and water (EPA 1996). Particles in air are usually divided to two categories, primary and secondary, based on the form in which they were emitted from their sources. Those emitted directly from sources as particles are called primary particles and those particles that are formed as the result of chemical reactions of gases emitted into the atmosphere, like SO_2 and NO_x, are called secondary particles. Both components are released from both natural sources and as a result of

anthropogenic activities. Natural sources for fine particles are mainly secondary sulphates arising from reactions of biogenic gases, volcanic SO_2, biogenic VOCs from organic matter and nitrates from NO_x. Anthropogenic emissions are typically released from energy production, traffic and from industrial processes (Seinfeld and Pandis 1998). In 1997, during the current study period, emission inventories in the Helsinki metropolitan area estimated that power plants emitted 55% (1100 t), traffic 39% (760 t) and other point sources 6% (110 t) of the total mass of 1970 tonnes of anthropogenic primary particles (Aarnio et al. 1998). Annual secondary precursor gas emissions for the same time period were much larger being 23800 t of NO_x and 8600 t of SO_2, respectively. Primary particles, SO_2, and NO_x emissions from traffic have all decreased consistently over the last ten years. Annual emissions of SO_2, NO_x and particles from power plants in 1997 represented reductions of 59%, 48% and 46% respectively from levels in 1990. In spite of a 1.5% increase in traffic volume from 1996 to 1997, overall emissions decreased due to a higher number of new cars using low emission technology, an increased number of catalytic converters in cars and decreased sulphur content of diesel fuel (Aarnio et al. 1998).

Recently, there has been a discussion among the scientists about the need to separate exposure to ambient and non-ambient particles because of regulatory and scientific reasons (Wilson et al. 2000, Mage 2001). Ambient concentrations are not correlating with exposure to total or non-ambient PM, but they are better correlated with exposure to ambient originated PM. Therefore, ambient concentrations are used in epidemiology as an estimate for exposure to ambient originated PM. Recently published results have shown different health effects for particles from different sources, which also suggests the need to separate exposure to ambient and non-ambient particles. Results reported by Laden et al. (2000) suggested that fine particles from combustion (traffic and coal burning), but not earth crust particles, were associated with increased mortality. Similarly, Pope et al. (2002) found that long-term exposure to fine particulate and sulphur oxide-related pollution were associated to cardiopulmonary and lung cancer mortality.

2.3.2 Emissions from indoor sources

The goal of most epidemiological studies has been to examine associations between acute health effects and exposure to ambient particles. Because people spend most of their time indoors (Jenkins et al. 1992, Clayton et al. 1993, Robinson et al. 1995, Jantunen et al. 1999),

the EE of particles from indoor sources is orders of magnitude higher than for outdoor sources and most of the exposure to ambient particles occurs indoors from ambient particles that have penetrated the indoor environment. Therefore, it is important to separate the PM generated by indoor sources from the PM infiltrated from ambient sources.

The U.S. EPA Particulate Total Exposure Assessment Methodology (PTEAM) study assessed exposures to PM_{10} of the non-smoking population of Riverside, CA (Clayton et al. 1991, Clayton et al. 1993, Thomas et al. 1993; Özkaynak et al. 1996). In addition to PM_{10} exposures, PM_{10} and $PM_{2.5}$ concentrations were monitored inside and outside of the home for the 178 randomly drawn study participants (age of 10-70 years). Gravimetric analysis of PM mass and chemical analysis of selected elements were carried out to identify the sources in these environments. Smoking, cooking, dusting and vacuuming were found to be the dominant indoor sources for high particle loads. PM concentrations in homes with ETS were considerably greater than those measured in homes without ETS. In homes with reported smoking particles of ambient origin, smoking, cooking and other sources contributed 60%, 30%, 3% and 7 % respectively to indoor $PM_{2.5}$ concentrations. Stepwise regression was carried out to identify major factors affecting indoor $PM_{2.5}$ concentrations. House volume and air exchange rate were found to be weak, but statistically significant determinants of residential indoor $PM_{2.5}$, in addition to outdoor air, smoking and cooking.

In addition to the PTEAM study, Wallace (1996) reviewed the results of the large-scale (greater than 150 homes) Harvard Six-City and the New York State ERDA studies together with several smaller studies in homes. All major studies found that the most important source of $PM_{2.5}$ was smoking, increasing indoor concentration 25 - 45 $\mu g/m^3$. Several studies found cooking the second important indoor source. It must be noted that none of the three major studies identified all indoor sources and thus a substantial unexplained portion of particle concentrations required identification in forthcoming studies.

One of the most important issues was to clarify the variability and contributions of ambient particles penetrating indoors. The PTEAM pilot study (Özkaynak et al. 1993) showed moderate correlation between outdoor and indoor $PM_{2.5}$ concentrations being higher ($r^2 = 0.54$) at night compared to during the day ($r^2 = 0.42$). Correlation of outdoor $PM_{2.5}$ bound sulphur with indoor $PM_{2.5}$ bound sulphur was extremely high ($r^2 = 0.9$). In the absence of indoor sources indoor/outdoor (I/O) ratio could be calculated from the equation: $I/O = a/(a+k)$, where

a is air exchange rate inside a residence and k is particle deposition rate (Wallace 1996). Deposition rates for $PM_{2.5}$ were determined to be about 0.4 1/h and a typical air exchange rate 0.75 1/h, thus at equilibrium the concentration of ambient $PM_{2.5}$ particles indoors would be 0.75/(0.75 + 0.4) = 65% of ambient concentrations.

A recent study by Abt et al. (2000) also reported cooking as an indoor source for fine particles. In addition, cleaning and people moving around the house produced increased concentrations of particles larger than 3 µm, demonstrating the role of re-suspension for coarse particles.

2.3.3 Source apportionment studies

The effects of particle sources on air quality can be analysed using receptor-modelling techniques in addition to providing information on emission inventories. Receptor models use air quality measurements of particle bound chemical species to apportion the contributions of various sources (Kao and Friedlander 1995). In this approach the main sources of particulate matter are resolved from the sample data measured in a certain location (the 'receptor'). The principles of receptor-modelling have been presented comprehensively by Gordon (1980), Cooper and Watson (1980), Henry et al. (1984), and Malm and Gebhart, (1996). Receptor models fall into several fundamental types: chemical mass balance methods, multivariate, microscopic and source-receptor hybrids. A widely used receptor model technique is the chemical mass balance (CMB) approach. The advantage of the CMB method is that huge data sets are not needed. The disadvantage of the method is that it requires information on chemical emission profiles from all significant sources, and contributions from sources that are not previously identified are not included.

Many multivariate methods are based on factor analysis (FA) of individual pollutants or elemental components to identify underlying patterns that explain common variations among a set of variables. Different variations of the FA techniques have been used to identify and quantify the sources of air pollutants (Kleinmann 1980, Henry et al. 1984, Morandi et al. 1987, Hosiokangas et al. 1999, Edwards et al. 2001). Principal component analysis (PCA) is one of the most common FA methods used to identify sources from air quality data (Thurston and Spengler 1985, Koutrakis and Spengler 1987). The main assumptions of PCA are: 1) the composition of the source emissions is constant, 2) the chemical species used in PCA do not react with each other and their concentrations are linearly additive, 3) the measurement errors

are random and uncorrelated, 4) the variability of concentrations between samples is dominated by changes in source contributions, 5) the effect of processes that affect all sources equally (e.g. atmospheric dispersion) is smaller than the effect of processes that influence individual sources (e.g. wind direction), 6) there are more samples than source types, and 7) the eigenvector rotations are physically meaningful (Seinfeld and Pandis 1998). In reality, discrepancies from these assumptions occur, but results from several studies have shown that despite of minor deviations from these assumptions, the main sources can usually be identified with PCA. For air quality data, let's say for *k* samples of *n* $PM_{2.5}$ bound elements, PCA is started by creating a correlation matrix for each pair of elemental concentrations. Each eigenvector of the matrix corresponds to a particular $PM_{2.5}$ composition influencing the receptor. Consequently, each sample collected at the receptor represents a linear combination of these eigenvectors. The corresponding eigenvalues can be used as a measure of the importance of each eigenvector for the receptor. Eigenvectors corresponding to eigenvalues greater than one, meaning that the eigenvector can explain more of the total variance of all variables than any single variable alone, are usually used for the rotation (Henry et al. 1984, Seinfeld and Pandis 1998). To clarify the meaning of the factors, the eigenvectors can be rotated. Often the remaining eigenvectors are rotated in such a way as to maximise the number of factor loadings that are close to one. The applied procedure is called Varimax rotation, where the factor vectors are all orthogonal i.e. they are independent of each other. In oblique rotation vectors can take any position in space (Kavouras et al. 2001). Thus, the rotated factors represent the major sources or meteorological effects, which explain the common variations in the elemental concentrations in PM samples. PCA is typically used instead of the CMB when source profiles are not available from all the sources contributing to the receptor, or the number of sources contributing to the receptor are not known. To apply PCA, quantitative source profile data are not needed, instead only the trace elements of the sources are used. PCA can identify the main sources, but it does not provide information on the source contributions to total PM concentrations. Thus, additional methods, such as multiple linear regression analysis (MLR), are needed. In MLR a trace element is needed for each source (i.e. factor identified in PCA). The trace elements are then used as independent variables in the MLR model (Henry et al. 1984). Commonly used trace elements are reviewed in Table 1.

Pakkanen et al. (2001b) analysed the composition of urban ambient $PM_{2.3}$ in Helsinki area from samples collected almost simultaneously during similar time periods to samples collected in the current study (1996-1997). The highest mass concentrations were reported for sulphate,

nitrate, soil, ammonium, and sea salt contributing 21%, 12%, 12%, 9% and 2.5% of the total $PM_{2.3}$ mass of 11.8 $\mu g/m^3$ respectively. A large portion of the average mass (43%) could not be identified with elemental and ion data. The authors concluded that the major contributors to the unidentified mass were carbonaceous compounds and water. Source apportionment of residential indoor $PM_{2.5}$ has been discussed in section 2.3.2. Source apportionment of exposure to $PM_{2.5}$, however, has been published neither in Finland nor in other countries.

Table 1. Summary of the trace elements used in source apportionment studies of PM.

Source category	Trace element / compound	Reference
Soil/crustal	Al, Ce, Fe, Mn, Sc, Si, Sm	Lee et al. 1994, Chan et al. 1991, Janssen et al. 1997, Huang et al. 1994, Maenhaut et al. 1989
Long-range transport, secondary particles	S	Gordon 1988, Van Borm et al. 1990, Janssen et al. 1997, APEG 1999
Tobacco smoke	Cd	Clayton et al. 1993
Road traffic	Br, C, Cu, Pb, Sb, Zn,	Gordon 1988, Harrison et al 1996, Kavouras et al 2001,
- catalyst cars	Al	Huang et al. 1994
- petrol vehicles	Al, Ca, Cd, Cu, Fe, Mn, Ni	Ojanen et al. 1998
- diesel vehicles	Al, Ba, Cu, Mg, Mn, Na, Pb, Sb, Zn	Chan et al. 1991, Ojanen et al. 1998
- non-catalyst vehicles	Br, Pb	Hildemann et al. 1991
- brake dust	Ba , Fe_2O_3, Mg^{2+}, SiO_2	Hildemann et al. 1991
- tire wear	Zn	Harrison et al. 1996
- road salt	Cl, Na	Ojanen et al. 1998, APEG, 1999
Subway	Mn	Crump 2000, Pellizzari et al. 2001
Sea salt	Cl, Na	Meanhaut et al. 1989, Van Born et al. 1990, Chan et al. 1991, Harrison et al. 1996, Janssen et al. 1997, APEG 1999
Steal industry/smelting	Ca, Cd, Cr, Cu, Fe, In Mn, Pb, Sn, Zn	Huang et al. 1994, Swietlicki et al 1996, Ojanen et al. 1998
- zinc smelter	Cd, Pb, Sn, Zn	Sweet et al. 1993
- copper smelter	Cu, P, Se	Sweet et al. 1993
- pyrite smelter	As, Cu	Pio et al. 1996
Refuse incineration	Ag, Cl, Cu, In, K, Pb, Sb, Zn	Van Borm et al. 1990, Sweet et al. 1993, Huang et al. 1994, Harrison et al. 1996, Parekh et al. 1987, Ojanen et al. 1998
Coal combustion	Ag, As, Cr, K, Mo, Pb, Sb, Se, S, Zn	Huang et al. 1994, Lee et al. 1994, Fung and Wong 1995, Parekh et al. 1987, Harrison et al. 1996, Biegalski et al. 1998, Ojanen et al. 1998
Oil combustion/refinery	Cr, La, Ni, Sm, S, V	Cass and McRae 1983, Van Borm et al. 1990, Lee et al. 1994, Huang et al. 1994, Harrison et al. 1996, Janssen et al. 1997, Ojanen et al. 1998, Hosiokangas et al. 1999
Wood/biomass burning	K, volatile C, elemental C	Chan et al. 1991, Huang et al. 1994, Janssen et al. 1997, Ojanen et al. 1998
Limestone/concrete	Ca, Mg	Huang et al. 1994

2.4 $PM_{2.5}$ concentrations

2.4.1 Ambient concentrations

Ambient air quality monitoring of $PM_{2.5}$ started during the mid 1980's in the United States and Canada. High urban $PM_{2.5}$ ambient concentrations were found in the Western United States in 1986-1994 showing 24-hour mean concentrations of 20 – 50 µg/m^3 (EPA 1996). The highest concentrations in the Eastern United States were found in industrial cities, such as Philadelphia and Pittsburgh (20 – 30 µg/m^3). Non-urban annual background $PM_{2.5}$ concentrations were 2-6 µg/m^3 in the Eastern United States and slightly lower in the Western parts of the country (1-5 µg/m^3). Concentrations in both of these areas were slightly higher in summer compared to winter (EPA 1996). Brook et al. (1997) reported 24-hour mean $PM_{2.5}$ concentrations of 13.9 µg/m^3 in 14 urban sites in Canada between 1986 and 1994.

Spatial distribution of the $PM_{2.5}$ concentration has been found to be relatively uniform across urban areas in several studies in the United States, Canada and Europe (Clayton 1993, Burton et al. 1996, Pellizzari et al. 1999, Oglesby et al. 2000a, Monn 2001, Houthuijs et al. 2001) suggesting that properly located ambient fixed site monitors may produce valid estimates for outdoor concentrations over large urban areas.

Continuous air quality monitoring of $PM_{2.5}$ started in Helsinki in March 1997. The mean annual urban concentrations of $PM_{2.5}$ have been 9 µg/m^3 (1997), 11 µg/m^3 (1998), 11 µg/m^3 (1999), and 8 µg/m^3 (2000) (Aarnio et al. 1998, Aarnio et al. 2001). Ruuskanen et al. (2001) reported similar low mean 24-hour ambient urban $PM_{2.5}$ concentrations in Helsinki, (9.4 µg/m^3), moderate concentrations in Alkmaar, The Netherlands (27 µg/m^3) and high concentrations in Erfurt, Germany (42 µg/m^3) during the winter of 1996-1997. Vallius et al. (2000) reported slightly lower 24-hour $PM_{2.5}$ ambient concentrations in Helsinki, during the winter of 1996-1997 (9.4 µg/m^3) than in spring 1997 (9.9 µg/m^3).

2.4.2 Microenvironment concentrations

Duan (1982) defined the term "microenvironment" as "a chunk of air space with homogenous pollutant concentrations". For example fine particle concentrations might be homogenous in the indoor air of all rooms in a residence (or metro, car, office, restaurant etc.) and could be defined as one single microenvironment. Another definition of microenvironment was

introduced by Mage (1985), which also took time into consideration. He defined microenvironment as a volume in space, during a certain time period, during which the variance of concentration in the volume is clearly less than the variance between this microenvironment and its surrounding microenvironments. Thus, in addition to fixed site ambient monitoring, exposures can be assessed by measuring the concentrations in indoor and outdoor microenvironments where people spend the majority of their time. This microenvironmental modelling approach offers a predictive tool to assess exposure if the time fractions spent in each microenvironment are known. Diaries and questionnaires are usually used to collect time-activity data (Monn 2001). Integrated exposure for an individual can be calculated as a time-weighed average of the concentrations of those microenvironments that he/she visited during the period. Thus, microenvironments, where a large proportion of time is spent and/or pollutant concentrations are high, must be taken into account. Residential outdoor and indoor, traffic, workplace, and other microenvironments like malls, restaurants, theatres and indoor sports halls have been considered important (Monn 2001). The microenvironment approach is much more demanding and expensive than ambient air monitoring, but it produces more reliable estimates of exposures. Therefore, this approach has been used in recent large-scale population exposure studies like PTEAM (Thomas et al. 1993), *EXPOLIS* (Jantunen et al. 1999) and the Toronto study (Pellizzari et al. 1999).

In the PTEAM study in California, mean 12-hour residential outdoor and indoor $PM_{2.5}$ concentrations were 48.9 $\mu g/m^3$ and 48.2 $\mu g/m^3$ showing high and similar levels in both environments (Clayton et al. 1993). Correlation between fixed site $PM_{2.5}$ and residential outdoor concentrations was high both during the day ($r = 0.83$) and during the night ($r = 0.93$). Indoor $PM_{2.5}$ concentrations correlated slightly less with ambient fixed site $PM_{2.5}$ concentrations. Exposure to $PM_{2.5}$ was not measured in this study, and thus could not be compared to residential or fixed site concentrations.

In the Toronto study (Pellizzari et al. 1999) mean 3-day residential outdoor and indoor $PM_{2.5}$ concentrations were 15.1 $\mu g/m^3$ and 21.1 $\mu g/m^3$ showing much lower concentrations than in PTEAM. The mean population exposure to $PM_{2.5}$, however, was higher (28.4 $\mu g/m^3$) than outdoor or indoor $PM_{2.5}$ concentrations. The correlation between residential outdoor and indoor $PM_{2.5}$ concentration was low, 0.32, and between residential outdoor concentration and personal exposure even lower, 0.23. In contrast, residential indoor concentrations and personal exposures to $PM_{2.5}$ were highly correlated with each other ($r = 0.79$). Low correlation between

outdoor and indoor or outdoor and personal exposure was concluded to be due to environmental tobacco smoke possibly present indoors.

Janssen et al. (1999) reported fine particle (PM_3) concentrations inside and outside a school in the Netherlands showing outdoor and indoor 24-hour mean concentrations of 16.8 µg/m^3 and 14.6 µg/m^3 respectively. The correlation between the classroom and the ambient concentrations was high (r = 0.96). In this study both longitudinal, repeated measurements on the same individuals, and cross sectional correlation, measurements of multiple individuals, were made. Mean longitudinal correlation between personal exposure and ambient fine particles was exceptionally high, 0.86 for all children and 0.92 for the non-ETS exposed children. The results of Janssen et al. (1999) suggest that ambient monitoring could be used to estimate daily variation of exposure to fine particles in epidemiological time series studies.

In Boston, the mean 12-hour indoor $PM_{2.5}$ concentration for individuals with chronic obstructive pulmonary disease (COPD) was higher than the outdoor concentration, 17.5 µg/m^3 and 14.2 µg/m^3 respectively (Rojas-Bracho et al. 2000). Individuals with COPD also had higher mean personal exposure, 21.6 µg/m^3, compared to indoor and outdoor concentrations. Indoor $PM_{2.5}$ correlated with outdoor concentration (r^2 = 0.49) moderately, but highly with personal exposure to $PM_{2.5}$ (r^2 = 0.60). The mean longitudinal correlation, based on repeated measurements of outdoor $PM_{2.5}$ and personal exposure measurements, was r^2 = 0.40, higher than in the earlier cross-sectional studies, but considerably lower than found for schoolchildren in Wageningen.

2.5 Exposure to $PM_{2.5}$

2.5.1 Special characteristics of the gravimetric analysis of the $PM_{2.5}$ exposure samples

Air quality guidelines and standards have been defined using mass concentrations. Therefore, personal monitors (PEM) also have been based on gravimetric analysis of PM collected on a filter. Gravimetric analysis of particles collected onto filters always has several sources of error. Battery operated PEMs are typically run at flow rates of 4 L/min or less, leading to net $PM_{2.5}$ masses as low as tens of micrograms per sample. The relative errors in PM concentration become significant, therefore, even when the absolute errors in weighing are as low as 5-10 µg.

To maintain high data quality in $PM_{2.5}$ exposure studies, sources of error in gravimetric analyses must be known and controlled during the weighing process.

A microbalance having reading precision of 1 µg or better (up to 0.1 µg are available) is needed when weighing $PM_{2.5}$ exposure filters. The precision of the mass analysed is not, however, as good as the precision of the balance reading, because external factors may affect the balance, the filter and the collected $PM_{2.5}$ mass. Typical factors controlled in weighing filters are relative humidity and temperature in the weighing room, thermal drafts, vibration of balance, and electrostatic charge (Lawless and Rodes 1999). Modern balances may eliminate the effects of temperature changes and vibration of the balance by computer-assisted algorithms. Static charge must be eliminated with external methods, usually by use of a radioactive polonium-210 neutraliser (Lawless and Rodes 1999, Allen et al. 1999). If uncontrolled, the static charge may cause weight discrepancies of ±50 – 500 µg (Lawless and Rodes 1999).

Carefully planned weighing procedures can also improve weighing precision. According to Allen et al. (1999) repeated weighing with 5-10 µg threshold to trigger a third weighing improves precision by a factor of 3. Subsequently, the effect of buoyancy on weighing results must be considered. When the density of the air changes from pre- to post-weighing, it also changes the observed weight of the filter. Allen et al. (1999) calculated that for a 37-mm Gelman™ Teflo filter (R2PJ037) the error caused by a 25 mmHg change in the air pressure is 3.5 – 4.5 µg. In practice for a single filter, the air pressure may change even more and thus lead to greater error in filter weight.

The selection of filter type is important in exposure monitoring. Currently, no filter material exists, which could be used for all desirable post-sampling analyses (Chartier and Weitz 1998). Filter material should also hold the collected particulate matter on the filter even when filters are moved during the sampling with the individual who is being monitored. In addition, low pressure-drop is an essential parameter to consider as PEM use batteries. Teflon filters have a low pressure-drop (Chartier and Weitz 1998) and are suitable, therefore, for exposure studies. Other advantages of Teflon filters are their low hygroscopicity and compatibility with post-sampling chemical analysis by X-ray fluorescence, ion coupled plasma and ion chromatography. Disadvantages of the Teflon filter are unsuitability for thermal carbon analysis.

Few studies have been reported dealing with the high quality gravimetric analysis of lightly loaded filters (Engelbrecht et al. 1980, Feeney et al. 1984, Chartier and Weitz 1998, Allen et al. 1999, Lawless and Rodes 1999). More studies are needed to identify and quantify the factors, which may significantly reduce the precision of the gravimetric exposure analysis. Especially interesting are of course errors, like the one caused by buoyancy, which can be corrected numerically from the weighing data.

2.5.2 Exposure studies

There are only a few large-scale population-based exposure studies reported in the literature for $PM_{2.5}$. Studies, of exposure to respirable suspended particles (RSP), often defined as $PM_{3.5 - 4.5}$, are reviewed together with $PM_{2.5}$ studies in the following section.

Dockery and Spengler (1981) reported one of the first exposure studies carried out in the United States. Non-occupational personal exposures of 37 volunteers to RSP were studied in Watertown MA and Steubenville OH. Study subjects carried personal samplers and filled time-activity diaries for 12 hours at a time. This general study design has been repeated in several later PM exposure studies. The main results of this study were that the 12-hour personal RSP exposure levels were in reasonably good agreement with mean outdoor RSP concentrations.

A little later, Sexton et al. (1984) reported RSP exposures of 48 volunteers in Waterbury Vermont. In this study residences were also monitored with indoor and outdoor microenvironmental RSP samplers. In contrast to the previous study, the main findings of this study were that outdoor particle concentration was not an important determinant of personal exposure, and average personal exposures were higher than indoor air levels, which were again higher than outdoor air levels.

Spengler et al. (1985) reported a RSP exposure study from two rural Tennessee communities in the United States. A total of 97 nonsmoking volunteers carried personal samplers, their homes were equipped with microenvironmental indoor monitors, and centrally located samplers in each of the towns monitored the ambient concentrations. Results from this study were that exposure concentrations of people who had been exposed to environmental tobacco smoke (ETS) were nearly twice as high as exposure of non-ETS exposed persons.

In the PTEAM study (Özkaynak et al. 1996) daytime personal PM_{10} exposure levels, as well as exposure levels of nearly all particle bound elements (except sulfur) were elevated relative to indoor and outdoor levels. For sulfur, which appeared to have no indoor or personal sources in Riverside, outdoor air concentrations explained 77 % of personal exposure concentrations.

Primary objectives of the Toronto study, with the highest number of measured personal $PM_{2.5}$ observations to date (n = 922), were to estimate the distribution of personal exposures of both smokers and non-smokers to manganese in $PM_{2.5}$ from the use of gasoline additive, MMT, in Canada (Pellizzari et al. 1999). It also contributed broader information on personal exposures to $PM_{2.5}$. Sampling was repeated for a new subset of the original sample on a monthly basis. This design allowed exposure assessments both across time and individual. In addition, assessments of exposure variation within groups, and both within and between individuals were assessed. The former is relevant for longitudinal, the latter for cross sectional studies. The study showed that cross sectional correlation of personal exposures to ambient air $PM_{2.5}$ was low. Results of correlation between individual exposures and ambient concentrations from repeated sampling have not yet been published.

Personal exposures of non-smokers to RSP ($PM_{3.5}$) were measured in several European cities to assess 24-hour exposures to environmental tobacco smoke (ETS). Participating cities in the study were Stockholm, Sweden (Phillips et al. 1996), Bremen, Germany (Phillips et al. 1998a), Prague, Czech Republic (Phillips et al. 1998b) and Basel, Switzerland (Phillips et al. 1999). In Stockholm a population of non-working men and women living in homes where residents actively smoked were exposed to 39 $\mu g/m^3$ RSP. In Bremen workers both living and working with smokers had the highest median 24-hour exposures to RSP reported in these studies, 789 $\mu g/m^3$. In Prague office workers living and working with smokers had the highest mean exposures to RSP, 60 $\mu g/m^3$, and housewives living in non-smoking homes had the lowest mean exposures 32 $\mu g/m^3$. Comparably, office workers living and working with smokers in Basel were exposed to 44 $\mu g/m^3$ RSP and housewives living in non-smoking homes were exposed to 31 $\mu g/m^3$. Clearly smoking was a significant contributor to RSP exposure in these studies.

Janssen et al. (1999) conducted an exposure study of 13 children in the Netherlands. Mean exposures to RSP of all children were 28.3 $\mu g/m^3$, composed of 24.4 $\mu g/m^3$ for children living

with non-smoking parents and 37.0 µg/m^3 for those living with parents that smoked. Personal exposures of participants were measured on several occasions, which allowed determinations of both individual regressions and cross sectional correlation between personal exposures and ambient air concentrations. Individual regression correlations between personal exposures to RSP and ambient air concentrations were considerably better than cross sectional correlations. Group exposures correlated extremely well with a median correlation coefficient of 0.92 for non-ETS exposed children.

Janssen et al. (2000) also studied $PM_{2.5}$ exposures of elderly non-smoking individuals with existing heart disease in Amsterdam and Helsinki, and the relationship between these exposures and ambient $PM_{2.5}$ concentrations. Exposure measurements were repeated biweekly 5 to 13 times for each participant. Median longitudinal Pearson correlation between personal $PM_{2.5}$ exposures and ambient $PM_{2.5}$ concentrations was high both in Helsinki and Amsterdam. In Helsinki, but not in Amsterdam, the mean $PM_{2.5}$ exposure level was lower than the mean ambient $PM_{2.5}$. Median exposures were below ambient concentrations in both Amsterdam and Helsinki. These results agree well with those of Sarnat et al. (2000) suggesting that personal $PM_{2.5}$ exposure levels for elderly, non-smoking individuals with sedentary lifestyles, are similar or below ambient levels, and correlation between personal exposures and ambient $PM_{2.5}$ concentrations for these individuals is high, but varies greatly between individuals and times of year.

Rojas-Bracho et al (2000) reported 12-hour personal, indoor, and outdoor $PM_{2.5}$, PM_{10}, and $PM_{2.5-10}$ exposures for 18 non-smoking individuals with chronic obstructive pulmonary disease (COPD) living in non-smoking residences in Boston, MA.Monitoring was performed for each participant for six consecutive days during the winters of 1996 or 1997 and for six to twelve days in the summer of 1996. Mean personal $PM_{2.5}$ exposures were higher than corresponding indoor and outdoor concentrations being 21.6 µg/m^3, 17.5 µg/m^3 and 14.2 µg/m^3 respectively. Higher personal exposures were associated with proximity of the individuals to indoor particle sources, such as cooking and cleaning. Indoor $PM_{2.5}$ concentrations were associated with both outdoor concentrations and personal exposures suggesting that home characteristics and indoor particulate sources were key determinants of personal-outdoor associations of $PM_{2.5}$ concentrations. Air exchange rates were also found to be important determinants of both indoor and personal levels. In addition, substantial interpersonal variability in the personal-outdoor relationship was reported.

Williams et al (2000) studied a population of elderly people (65+ years of age) living at a retirement centre in Baltimore, USA, as a sensitive population to $PM_{2.5}$ exposures. Mean 24-hour exposures, residential indoor and outdoor $PM_{2.5}$ concentrations were 12.9 $\mu g/m^3$, 9.4 $\mu g/m^3$ and 22.0 $\mu g/m^3$ respectively. This result suggested that exposures to $PM_{2.5}$ were still higher than indoor concentrations even though the activity level of elderly people might be lower compared to working populations. There were also much lower $PM_{2.5}$ concentrations indoors compared to outdoor air showing that building shells reduce indoor exposures to particles from outdoor sources.

In summary, it is clear that the most important activity to increase exposures to $PM_{2.5}$ is smoking. Other significant factors identified were outdoor air, cooking, kerosene heaters and wood burning. It is clear from a review of the literature, however, that more detailed analyses of determinants of $PM_{2.5}$ exposures are needed including clarification of relationships between $PM_{2.5}$ sources, exposures and health effects. This thesis addresses some of these gaps in information and, in particular, provides detailed information on how exposures to $PM_{2.5}$ are linked to sources. In addition, as limits for air quality and monitoring in many nations is assessed using ambient fixed site measurements, this thesis examines the validity of ambient air monitoring to estimate source related exposures.

3 AIMS OF THE STUDY

The aims of the current study were to answer the following questions:

1 What is the precision of personal and microenvironment $PM_{2.5}$ concentration measurements, and how comparable are the two sampling methods?

2 What are personal exposures of adult urban populations to $PM_{2.5}$?

3 What are the determinants of personal exposures to $PM_{2.5}$ in adult urban population?

4 What are the main source categories of $PM_{2.5}$ and their contributions to personal exposures in adult urban population?

5 What is the applicability of ambient monitoring for estimation of personal exposures to $PM_{2.5}$ from different sources?

4 MATERIALS AND METHODS

4.1 Study design

This thesis is based on fieldwork carried out from fall 1996 to fall 1997 as a part of the European multi-centre air pollution exposure study *EXPOLIS* (Jantunen et al. 1998, Jantunen et al. 1999). Microenvironment and personal exposure monitoring was performed over 48 hours on 201 randomly selected adults (age between 22 and 55 years) to assess $PM_{2.5}$ exposure distributions of the population in the Helsinki Metropolitan Area. Time-microenvironment-activity diaries (TMAD) and questionnaires were used to collect background information for each study subject. A pilot phase was carried out to develop and test equipment and procedures for the actual field phase. Quality assurance and quality control (QA/QC) procedures were carefully planned and tested to maximise the quality of the collected data (see section 4.6).

The original idea of the study was to sample each individual 3 times, but after summarising scientific and financial realities, only one measurement per study subject was possible. The advantages of repeated measurements had to be balanced against the disadvantages of very small sample sizes and consequently the representative population samples were considered more important than the ability to compare within and between individual differences (Jantunen et al. 1999).

4.2 Population sampling and study location

It is essential to assess the extent to which the study population was representative of the population in the Helsinki metropolitan area before results can be generalised. These analyses for the *EXPOLIS* study have been presented in papers published by Rotko et al. (2000) and Oglesby et al. (2000b). In Helsinki a Short Questionnaire was mailed to a random *Base* sample of 2523 individuals drawn from the census, and 1882 *Respondents* returned a valid questionnaire attaining the targeted response rate of 75%. Two sub-samples from these *Respondents* were randomly selected: 1) 234 *Diary* subjects for 48-hour time-microenvironment-activity diary (TMAD) and extensive exposure questionnaires, and 2) 201 Exposure subjects, similar to *Diary* subjects plus 48-hour personal exposure and microenvironment monitoring.

Bias in the sample population was assessed by comparison of *Respondents* with the total population of Helsinki (Rotko et al. 2000). The share of women and individuals with more than 14 years of education was higher among *Respondents* than in the overall population. Men, participants between 25-34 years of age and unmarried individuals were somewhat under represented in the study population in Helsinki. The two sub-samples differed from the *Respondents* in having more employed and higher-educated individuals, however, the differences were not remarkable. Somewhat surprisingly, the largest sample bias was reported at the first and easiest step of responding to the mailed Short Questionnaire, and not at the last and most demanding stage of participating in the exposure measurements. According to Rotko et al. (2000) the *Diary* and *Exposure* subjects in Helsinki are fairly representative of the larger population, and selection bias would not overly impact analyses of predictors of personal exposures or analyses within a city.

The possibility that subjects would have altered their behaviour on days of personal sampling (i.e. those who carried the PEM case) was assessed by using time-activity data. Those who were monitored spent slightly more time indoors at home (median time 26h 34 min) compared to those who were not monitored, but filled the time-activity–diaries (25h 43 min) during 48 hour sampling period (Jantunen et al. 1999). This difference may not lead to a significant bias in behaviour of the study subjects caused by monitoring their exposure to $PM_{2.5}$.

Helsinki (http:/www.hel.fi) is the capital of Finland and is located on the southern coast on the Gulf of Finland. The population of Helsinki metropolitan area is about 1 million. A great majority of the population works in offices and services, but engineering, electronics, shipbuilding, food processing and timber industries are also important for the economy of the area. Climate exhibits both maritime and continental influences. Surrounding seas cool the climate in spring but warm it in fall. The sea is frozen and the ground covered with snow for a extended period in winter. Mean temperature in January is -6 °C, but the summer months are mild (17 °C). Prevailing south-westerly sea winds often dilute local emissions leading to low air pollution levels. National air quality guidelines as well as respective EU standards for PM_{10} are commonly exceeded, however, during the spring dust episodes as a result of the melting of the snow and ice-fall from the winter, releasing the gravel that was spread for car traction. NO_2 sometimes exceeds air quality guidelines in winter during extended inversions (Aarnio et al. 1998, Kukkonen et al. 1999).

4.3 Monitoring methods of $PM_{2.5}$ (I)

4.3.1 Personal exposure monitoring

A personal exposure monitor (PEM) was developed for *EXPOLIS* to measure $PM_{2.5}$ exposures (see publication I). The PEM consisted of a pump, a cyclone and a filter holder packed into an aluminium briefcase together with a CO monitor, VOC sampling tube and battery pack. The modified Buck IH pump (A.P.Buck Inc. Orlando, FL, USA) was capable for sampling for over 48 hours with one set of batteries (6 industrial strength alkaline D-cells) and was therefore suitable for personal measurements. The air samples were pumped at a flow rate of 4 L/min through a small $PM_{2.5}$ cyclone (GK2.05 KTL Cyclone) and a 37 mm Millipore filter holder (Millipore Corporation, Bedford, MA, USA) with Gelman Teflo™ filter (2 µm pore size having a collection efficiency of 99.99 % for 0.3 µm particles). The design and performance of the GK cyclone family is reported by Kenny and Gussman (1997). The KTL cyclone used in this study was designed specifically for *EXPOLIS* by BGI Inc (Waltham, MA, USA). Teflon filters were chosen, because of high collection efficiency, low chemical background and low pressure-drop needed for PEM sampling.

Each subject was monitored for 48 hours. Two consecutive days were monitored to collect enough mass for reliable concentration results. Two filters were provided for each subject. One 'day filter' for the two periods beginning with the participant leaving home for work and ending with the participant returning home from work (about 2 x 8-10 hours), and one 'night filter' for the remaining times (about 2 x 14-16 hours). The subjects were personally instructed about the correct procedure for changing the (day or night) filters during the study period. The $PM_{2.5}$ methodology, QA/QC and pilot results are described in publication number I.

4.3.2 Microenvironment monitoring

A microenvironmental monitor (MEM) was also constructed for the *EXPOLIS* study (see publication I). The MEM sampler contains an EPA-WINS impactor (EPA Well Impactor Ninety-Six, BGI), a filter holder with a 47-mm Gelman Teflo™ filter and a PQ100 pump (BGI). The EPA-WINS impactor was selected as a federal reference method (FRM) sampler for $PM_{2.5}$ in the USA (EPA 1997). It is designed to remove 50% of particles with an aerodynamic particle diameter of 2.5 µm at a flow rate of 16.7 L/min prior to collection on the

filter. The pump could be programmed to start and stop sampling automatically, and used mains power supply (220/110V AC) or a 6 V internal lead acid battery.

In the field procedure residential indoor and outdoor and workplace monitoring was performed with MEMs. The pumps were programmed to run in the residences of the subjects for the expected non-working hours and in the workplace for the expected working hours of each subject during the 48-hour monitoring time. Flow rates were calibrated before and after each sampling with a bubble flow meter (e.g. Buck M-30) as a primary standard.

Sample volumes measured by PEMs were normalised to 101.3 kPa (760 mmHg) air pressure at 20 °C, the conditions in which the MEMs were calibrated, because PEM pumps had volumetric flow control and MEM pumps mass flow control. The PEM flows were normalised using the temperature data collected by the external temperature sensor of the Langan CO monitor (Langan Products Inc., San Francisco, CA, USA) that was also present in the PEM aluminium briefcase. Air pressure data needed for the normalisation of PEM volumes were measured in the weighing room by a mercury manometer.

4.3.3 Filter weighing (I, II)

The need for high-quality gravimetric analysis increases when weighing filters with small masses of particulates that are typical of many exposure studies. The precision of the weighing procedure must be considerably below 10 µg and typical analytical balances with the precision of 10 µg or more used in weighing filters were not sufficient. In addition, stable weighing room conditions and careful weighing procedures are required.

In *EXPOLIS*-Helsinki a microbalance with the reading precision of 1 µg (Mettler MT5 by Mettler-Toledo AG, Greifensee, Switzerland) was used for weighing the Teflon filters. Filters were equilibrated to weighing room conditions for a minimum of 16 hours before and a maximum of 36 hours after the sampling period. Temperature, relative humidity and air pressure were recorded during each weighing session. For stability the weighing room was located on ground floor, had no external walls or windows and minimal air exchange. An electrostatic air cleaner (Elixair model E400C1) was operated in the weighing room to keep the air as clean as possible.

In the weighing procedure, two consecutive weightings of the same filter had to agree within 1 µg to be recorded. If the second measurement differed more than 1 µg from the first, both were rejected, and the procedure was repeated until the ±1 µg goal was met.

Teflon filters are electrically non-conductive and therefore easily acquire static charge. Such charge strongly affects the gravimetric analysis if present. Each filter was discharged, therefore, before weighing using a Polonium-210 alpha radiation source (500 µCi Staticmaster 1269 by Cahn Inc. USA) by moving the filter near the source.

To control for weighing conditions and balance stability, three 37 mm and three 47 mm laboratory blank filters were weighed in each weighing session. The microbalance calibrated itself automatically, when the balance detected the need for internal calibration. In addition to laboratory blanks, an external 200-mg stainless steel standard weight was weighed at the beginning and at the end of each weighing session.

When the time series of laboratory blank filter masses was analysed, a strong correlation between the observed mass and the air pressure in the weighing room was noted. The reason for the effect was identified as the variation in air buoyancy. This phenomenon has been known for a long time, but typically has been neglected in filter weighing QA procedures. In this study, we wanted to assess if this phenomenon should be taken in to account. The equations and the overall procedure to calculate the buoyancy effect when weighing filters are presented in paper I. The effect of buoyancy under the conditions for the one-year field study was first analysed by calculating possible mass changes caused by this effect. The analysis showed, that the buoyancy effect can be up to 10 – 15 µg, and therefore is an important, but correctable source of error. Therefore, air buoyancy effect was corrected from all the weighing results throughout the study. The correction ranged from –7 to +13 µg.

Analysis of the significance of the air buoyancy effect among other sources of error in weighing blank filters is presented in paper II. Other potential sources of error include temperature, relative humidity and static charge and are well known and frequently controlled. Quantitative analysis of the sources of error, air buoyancy variation, filter storage time, relative humidity, balance random variation and filter handling, showed that, after eliminating the static charge the most important correctable factors were air buoyancy variation and filter storage time explaining 35% of the variation in uncorrected data. The standard deviation of the

differential blank filter weighing after applying all corrections was 2.7 µg. In addition to the errors quantified for the blank filters, other factors that also should be considered when sampling PM are hygroscopicity of the particulate material collected on the filters (e.g. sulfates and salts that contain variable amounts of water, e.g. Lipfert 1994, Hämeri et al. 2000) and volatilisation of sampled material (e.g. nitrates, Lipfert 1994, Hering and Cass 1999).

4.4 Elemental and black smoke analysis of the $PM_{2.5}$ samples

Elemental analysis of the 499 filters was performed by energy-dispersive X-ray fluorescence spectrometry (ED-XRF) using X-Lab 2000 (SPECTRO Analytical Instruments, Germany 1998) in the University of Basel, Institute for Mineralogy and Petrography (Switzerland). The ED-XRF method was chosen, because of advantages in analysing large sets of exposure samples compared to other methods, applicability for analysing particulate bound elements on untreated Teflon filters, relative precision within 10%, multi-elemental capability and detection limits in the range of a few ng/cm^2 in a 40-minute analysis per sample. An important additional factor was that ED-XRF sampling was non-destructive and the filters and particulate mass remain intact and can be used for further investigations. With this method elements with an atomic number between Z = 11 (Na) and 92 (U) can be detected. Detailed description of the elemental analysis of the $PM_{2.5}$ samples is presented in Mathys et al. (2001a).

The limit of detection (LOD) was calculated individually for each element in each sample, as concentrations of eachelement in ED-XRF often affects the detection of another element. This means that there was a distribution of LODs for each element rather than a single value. The lowest LODs were found for Ga, below 1 ng/cm^2, and the highest for Na, exceeding 100 ng/cm^2. The LODs for all the analysed elements are presented in Mathys et al. (2001a). The relative standard deviation of the duplicates for elemental analysis ranged from 3.4% (Calcium) to 4.8% (Bromine) (Oglesby et al. 2000a).

The expression black smoke (BS) is used in this thesis synonymously to indicate the absorption of $PM_{2.5}$ samples measured using a reflectometer (EEL model 43, Diffusion Systems Ltd., London) according to the method defined in the international standard ISO 9835 (ISO 1993) applying the OECD protocol (OECD 1964), with modification of particle cut-size and filter material.

4.5 Source apportionment

Principal component analysis (PCA) with Varimax rotation was applied to identify sources from the exposure and the microenvironment samples. Natural log-transformed elemental and Black Smoke concentrations were used for the PCA. Elemental concentrations below detection limit were replaced by half the detection limit. The PCA methodology is also described in section 2.3.3 and in paper V.

Mass contributions of sources identified in the PCA analysis were determined using source reconstruction (SR). In SR elemental concentrations were combined with source fingerprint or trace element data for the main source categories. The SR method is described in detail in paper V.

4.6 Quality assurance (QA)

In an international multi-centre study like *EXPOLIS,* special attention must be paid to quality assurance (QA) and quality control (QC) issues to get comparable data between different study centres. An intensive six-month long pilot phase was carried out in Helsinki, therefore, ending in a one-week training workshop for *EXPOLIS* team members from all centres. In the pilot phase, sampling monitors and weighing procedures were tested and finalised prior to the actual field phase. The standardized procedures were then distributed to all study centres in the form of detailed standard operating procedures (SOP). Four SOPs were created for $PM_{2.5}$ sampling and analysis (see paper I). Thus, QA protocol was established and the QA Unit of KTL-National Public Health Institute evaluated procedures according to these protocols several times during the fieldwork.

The detection limits for $PM_{2.5}$ after the pilot phase, were 1.0 $\mu g/m^3$ and 2.8 $\mu g/m^3$ for MEM and PEM respectively. Precision of the $PM_{2.5}$ concentrations for PEM and MEM were assessed as relative standard deviations (RSD) in a similar manner to those reported by Thomas et al. (1993) for the PTEAM study. In this study median RSDs for PEM and MEM were 6.7% and 2.3 % respectively. The accuracy of the $PM_{2.5}$ concentrations could not be determined, because 'correct' reference values were not available. Instead the accuracy of the airflow was checked before and after each sampling with bubble flow meter as a primary standard (Buck M-30 by A.P.Buck Inc. Orlando, FL, USA). QA objectives for PEM flow rate before sampling had to be

within 2.5% and after sampling within 10% of the 4 L/min target value. For MEM, the normalised flow rate before and after measurement period had to be within 3% of the target flow rate of 16.7 L/min. Every sample met these goals. Accuracy of the balance was tested in an accredited calibration laboratory (G.W. Berg Inc.) before and after the study. It was found to be excellent and not changed during the study.

Comparability of the PEM and MEM $PM_{2.5}$ results were assessed in a side-by-side test of 5 PEMs and 10 MEMs showing precision of 3.0% as mean RSD. The $PM_{2.5}$ measurement methodology, quality assurance procedures and pilot results of the *EXPOLIS* study are presented in detail in paper I.

4.7 Data analysis

Wilcoxon, Kolmogorov-Smirnov, t-tests and linear regression analyses in paper II were analysed using STATA version 5.0 (Stata Corporation, College Station, Texas, USA). All other statistical tests and analyses presented in this thesis were computed with Statistical Package for Social Sciences (SPSS for Windows, SPSS Inc. Chicago, Illinois, USA) version 9. Statistical tests and analyses in the publications are summarised in Table 2.

Table 2. Statistical tests and data analyses.

Paper no.	Test or analysis used	Purpose of analysis
II, III	Linear regression [1]	To identify the factors that produced variation for blank filters Relationship between expposure and one or more predictor variables
III	Analysis of variance (ANOVA)	Variability of exposure between categorised sub-populations
III, IV	Wilcoxon test Kolmogorov-Smirnov test	Difference between exposed sub-populations
IV	t-test	Difference between exposed sub-populations
V	Principal Component Analysis (PCA)	To identify main source categories from exposure and microenvironmental samples

[1] Ln-transformed data used for exposure and microenvironmental concentrations

5 RESULTS

5.1 $PM_{2.5}$ exposures

Descriptive statistics of personal weekday exposures, microenvironment and ambient fixed site $PM_{2.5}$ concentrations for the whole study population, including those exposed to environmental tobacco smoke (ETS) and those not exposed to ETS, are presented in Table 3. Subsequently, these concentrations were stratified by behavioural, microenvironment and socio-demographic factors and are presented in the following chapters.

Table 3. Summary statistics of personal 48-hour exposures (Exp_{48h}) to $PM_{2.5}$, exposures during commuting and working hours (Exp_{work}), exposures during non-working hours ($Exp_{leisure}$), and $PM_{2.5}$ concentrations in residential indoor (Ind), workplace (Wrk), and residential outdoor (Out) microenvironments. Central ambient fixed site $PM_{2.5}$ concentrations (Amb) are shown for the same time periods as the residential outdoor samples.

	n	AM	GM	GSD	50%	75%	95%	Max
Exp_{48h}	194	15.4	10.9	2.13	9.93	17.6	43.0	177
Exp_{work}	194	18.8	11.5	2.44	10.5	18.7	57.2	234
$Exp_{leisure}$	196	12.7	8.94	2.15	7.74	14.9	30.8	147
Ind	192	12.0	8.43	2.14	8.09	12.8	28.8	119
Wrk	151	15.7	7.82	2.78	7.08	14.4	50.3	278
Out	170	9.55	7.70	1.92	7.32	12.7	22.8	44.1
Amb	96	10.1	8.55	1.81	8.53	13.7	20.6	29.7

AM = arithmetic mean, Std = standard deviation, GM = geometric mean,
GSD = geometric standard deviation, 50%, 75%, 95% ith percentile, Max = maximum value

Highest mean exposures for the whole adult population were found during working hours (18.8 $\mu g/m^3$). The mean private time exposure (12.7 $\mu g/m^3$) was slightly higher than mean residential indoor concentrations (12.0 $\mu g/m^3$), which again exceeded mean residential outdoor air concentrations (9.55 $\mu g/m^3$). The Helsinki Metropolitan Area Council initiated ambient fixed site $PM_{2.5}$ monitoring part way through sampling for this study. The number of samples for ambient fixed site (Amb) was lower, therefore, than the number of residential outdoor samples collected in this study. Linear regression analysis of residential outdoor and ambient fixed site concentrations showed high spatial homogeneity (see paper III, residential outdoor $PM_{2.5}$ = 0.97 x fixed site $PM_{2.5}$ - 0.50, $r^2 = 0.91$, $p = 0.000$) suggesting that fixed site monitoring can be used to predict residential outdoor concentrations for $PM_{2.5}$. The largest variations in the $PM_{2.5}$

concentrations were found in workplaces, both in microenvironment and exposure samples. The lowest variation was found in ambient air. Maximum and 95th percentile values showed a similar pattern, having the highest concentrations in workplaces and during personal daytime activities.

5.2 Determinants of personal exposures to $PM_{2.5}$ (III-IV)

5.2.1 Behavioural and microenvironmental determinants (III)

Smoking was the strongest determinant of the 48-hour $PM_{2.5}$ personal exposure (III; Table 1). Smokers were exposed to almost 3 times higher median $PM_{2.5}$ concentrations in their immediate environment, not including active smoke inhalation from the cigarette into their lungs, compared to participants not exposed to environmental tobacco smoke (ETS). Non-smokers who were exposed to ETS in any environment had about 2 times higher median exposures than those not exposed to ETS.

Residential indoor concentrations were also almost 2 times higher in those residences where someone smoked indoors (III; Table 1). Smoking was also associated with higher workplace concentrations, but the difference was not statistically significant, probably due to the fact that smoking in the workplace is prohibited in Finland, and also due to the large variation in $PM_{2.5}$ workplace concentrations.

In general, $PM_{2.5}$ exposures were higher than residential and workplace concentrations both for the ETS exposed and non-exposed participants (III; Table 1). Residential indoor concentrations were higher than ambient levels in those homes where someone smoked indoors. In contrast, residential indoor concentrations were lower than outdoor concentrations in non-smoking homes.

When simple linear regression was used to predict $PM_{2.5}$ 48-hour exposures for all participants of *EXPOLIS*–Helsinki using microenvironment $PM_{2.5}$ concentrations as independent variables, residential indoor and workplace concentrations were the best predictors explaining 53% and 38% of the variation in personal exposures respectively (III; Table 2a). In contrast, residential outdoor and ambient fixed site concentrations could explain only 16% and 17% of personal exposures respectively. When similar analyses were performed for non-ETS exposed

participants, residential indoor $PM_{2.5}$, workplace $PM_{2.5}$, ambient fixed site $PM_{2.5}$ and residential outdoor $PM_{2.5}$ concentrations explained 46%, 46%, 41% and 39% of the variation in personal exposures respectively.

Multiple linear regression was also performed to predict 48-hour $PM_{2.5}$ exposure for the non-ETS exposed participants (III; Table 2b). Predictors identified in stepwise regression were $PM_{2.5}$ residential indoor concentration, $PM_{2.5}$ workplace concentration and traffic density of automobiles and light-duty vehicles on the street nearest the home. This model explained 77% of the variance of personal exposure. Candidate variables included in the analysis but excluded by the step-wise regression were: residential outdoor $PM_{2.5}$ concentrations, density of truck traffic near the home, home to work distance, time windows were kept open at home, time spent in the following activities; cooking, walking, using motorbike, in car, in bus (also combined car and bus) and in train. In addition, the following categorised variables were used: vacuum cleaning during the sample period (yes-no), home location (suburban single home, suburban high rise, downtown), work location (suburban, downtown), heating season (winter, summer), time of year (spring, summer, fall, winter) and stove type (electric, other). More detailed description of the variables is presented in paper III; Table 2b.

Subsequently, similar regression analysis was applied without residential indoor $PM_{2.5}$ concentration and $PM_{2.5}$ workplace concentration as candidate variables. This analysis demonstrates the ability to predict personal exposures using frequently available ambient concentration data and additional information, which can be collected by questionnaires or is available from established databases (e.g. census), but without microenvironment measurements. Predictors of personal exposures for the non-ETS exposed participants were residential outdoor concentration and home location, but the model accounted for only 47% of the variance of personal exposures.

Some categorised variables were also analysed to identify sub-groups and periods in which personal exposures to $PM_{2.5}$ were significantly elevated above the rest of the population (III; Table 3). $PM_{2.5}$ 48-hour exposures were higher in summer compared to other seasons. In addition, elevated exposures were found when windows were open at home more than 30 hours during the 48-hour study period or when participants lived in downtown compared to sub-urban neighbourhoods.

5.2.2 Socio-demographic determinants (IV)

$PM_{2.5}$ exposure distributions and microenvironment concentrations stratified by socio-demographic variables have been presented in paper IV. For the whole study population the two highest 48-hour exposures were found among unemployed men and among men with low occupational status (41.8 µg/m^3 and 26.1 µg/m^3 respectively, IV; Table 6). These high exposures mainly reflected personal daytime exposures, where the two highest exposures were also found among the unemployed and participants with low occupational status (30.4 µg/m^3 and 30.2 µg/m^3 respectively, IV; Table 3). Private time $PM_{2.5}$ exposures were also highest among participants with low occupational status and in single adult families (14.2 µg/m^3 and 14.1 µg/m^3 respectively, IV; Table 4).

Exposures to $PM_{2.5}$ for those who were not exposed to ETS were lower in all socio-demographic groups compared to ETS exposed participants in the same socio-demographic groups (see IV; Tables 2 and 5). Education and occupational status were also significant predictors for exposures to $PM_{2.5}$ for both non-ETS exposed participants and for the whole population.

The three highest mean residential indoor $PM_{2.5}$ concentrations were found in families with one adult, among women, and among young people (14.3 µg/m^3, 14.2 µg/m^3, and 13.6 µg/m^3 respectively, IV; Table 4). Maybe surprisingly, men were exposed to the lowest mean indoor concentration (9.3 µg/m^3).

People with low occupational status and those with less than 14 years of education had two highest workplace $PM_{2.5}$ concentrations (32.8 µg/m^3 and 28.6 µg/m^3 respectively, IV; Table 3). In general, variation in workplace $PM_{2.5}$ concentration was larger than in any other microenvironment. The highest 95th percentile concentrations (185 µg/m^3) were found in populations with low occupational status and among those with less than 14 years of education.

There were no big differences in $PM_{2.5}$ residential outdoor concentrations between socio-demographic sub-populations (IV; Table 4). The highest mean concentrations were found in a population of women and among those with low occupational status (10.4 µg/m^3 and 10.3 µg/m^3 respectively). Men lived in locations where the mean residential outdoor concentration (9.0 µg/m^3) was the lowest compared to other sub-populations.

Only years of education were included in multiple linear regression models to predict exposure to $PM_{2.5}$ using socio-demographic factors as candidate variables ($p= 0.011$, $r^2 = 0.05$, $n = 126$). The following candidate variables were included in the stepwise analysis, but excluded from the model: gender, age, occupational status, employment, number of adults at home and number of children at home (see detailed descriptions of the variables from IV; Table 2).

Only residential indoor and workplace concentrations were identified as predictors of 48-hour exposure for the non-ETS population ($p= 0.000$, $r^2 = 0.70$, $n = 83$), when socio-demographic factors listed above were used as candidate variables in addition to residential indoor, outdoor, and workplace $PM_{2.5}$ concentrations.

When all behavioural, environmental (see III; Table 2b) and socio-demographic factors were included as candidate variables in multiple regression, socio-demographic variables were not identified as significant predictors of exposure. Predictors of 48-hour exposures were residential indoor and workplace concentrations and traffic density of automobiles and light-duty vehicles on the street nearest the home, as in the corresponding analysis without socio-demographic variables (III; Table 2a). Socio-demographic descriptors, however, were included into the model, when similar analysis was performed without residential and workplace concentrations. In this model residential outdoor $PM_{2.5}$ concentration, years of education, age and traffic density on the street nearest the home were predictors of 48-hour exposure ($p= 0.000$, $r^2 = 0.55$, $n = 85$).

5.3 Source apportionment (V)

5.3.1 Source identification

The strongest source for $PM_{2.5}$ personal exposure was smoking (see section 5.2.1). It was also clearly associated with increased indoor $PM_{2.5}$ concentrations in residences and workplaces.

Other main source categories were identified using principal component analysis on $PM_{2.5}$ elemental and black smoke concentrations. Elemental composition of $PM_{2.5}$ samples collected in the *EXPOLIS* study was reported by Mathys et al. (2001b). The main source categories identified from exposure and microenvironment samples for non-ETS exposed participants are summarised in paper V; Table 5. The sources of ambient origin - long-range transport primary

and secondary particles, particles emitted directly from local combustion sources, soil dust, and sea and de-icing salt - were found in all microenvironments. In addition to these source categorises, an indoor source, representing detergents and possibly cooking, was identified from indoor environments.

Analysis of the median indoor/outdoor (I/O) ratios showed that there were no or few indoor sources for lead, zinc, sulphur, bromine and black smoke (V; Table 6). These elements are typically related to particles from traffic and long–range transport. In contrast, median I/O ratios for silica, aluminium, chlorine, and potassium were above 1 suggesting both indoor and outdoor sources for these elements. Therefore the analysis of the I/O ratios confirmed the source identification by PCA.

5.3.2 Contributions of source categories to $PM_{2.5}$

Tobacco smoke was the largest contributor to $PM_{2.5}$ exposures of non-smoking participants exposed to ETS and contributed 6.70 µg/m^3, or 40% of total exposure. ETS provided even higher contributions to the exposure of participants that smoked, contributing 21.1 µg/m^3, or 68% of the total exposure.

Contributions of other sources to $PM_{2.5}$ microenvironment concentrations and exposure are presented in paper V. Inorganic secondary PM had the highest contribution to ambient $PM_{2.5}$ concentrations, contributing 4.7 µg/m^3 (V; Figure 1). Indoor concentrations of secondary PM in residences and in workplaces were about 70% of the ambient levels, being 3.3 µg/m^3 and 3.4 µg/m^3, respectively. Personal exposures showed similar average contributions as residential and indoor microenvironments with 2.9 µg/m^3. All distributions were lognormal. The range of secondary PM concentrations was larger in ambient air compared to personal exposures, 0.383 - 14.1 µg/m^3 and 0.306 – 8.93 µg/m^3 respectively.

The source category ‘Other particles’ including primary combustion particles, non-volatile organic particles, particles from tyre wear, etc. (for short, combustion and other particulate matter, elsewhere in this thesis ‘CoPM’ is used to refer to this category) had the second highest contribution to ambient $PM_{2.5}$ concentrations, contributing 3.5 µg/m^3. Indoor concentrations of CoPM in residences and workplaces were 74% and 83% of ambient levels. Personal exposures to CoPM were lower than ambient concentrations, but slightly higher compared to indoor

concentrations, contributing 3.0 µg/m^3. Personal exposure/ambient ratio for CoPM (0.86) was clearly higher than that for secondary PM. Thus, exposures to CoPM reflected concentrations in those microenvironments where people spent most of their time, i.e. home indoors and in workplaces.

Soil dust was the third major contributor to ambient $PM_{2.5}$. In contrast to secondary PM and CoPM, the mean ambient soil dust concentration was lower, 1.6 µg/m^3, compared to residential indoor, workplace and exposure concentrations, contributing 2.5 µg/m^3, 2.4 µg/m^3, and 2.5 µg/m^3, respectively. Clearly there is also an indoor source present for soil dust, which is supported by the larger concentration range found in residential indoor air, 0.247 µg/m^3 – 8.56 µg/m^3, than in residential outdoor air, 0.124 – 5.96 µg/m^3. Personal exposures to soil dust ranged between 0.482 µg/m^3 and 7.55 µg/m^3.

In addition to the three major contributors to ambient $PM_{2.5}$, one minor contributor, sea salt, was identified with PCA and source reconstruction. Mean residential outdoor sea salt concentrations, 0.297 µg/m^3, were higher than mean indoor concentrations in residences and workplaces, contributing 0.229 µg/m^3 and 0.128 µm respectively. Mean personal exposures to sea salt, 0.202 µg/m^3, were between outdoor and indoor concentration levels.

In addition to the major source categories identified in indoor and personal exposure environments, there was also a minor contributor, detergents, to $PM_{2.5}$ concentrations. Based on a fairly small number of quite high concentrations mean personal exposures were 0.580 µg/m^3, and mean residential indoor concentrations were 0.539 µg/m^3, both much higher than workplace concentration of 0.2 µg/m^3.

5.4 Applicability of ambient and indoor monitoring for personal exposure assessment to $PM_{2.5}$ from different sources (III, V)

Table 4 shows the results of simple linear regressions of 48-hour $PM_{2.5}$ exposure (Exp_$PM_{2.5}$), residential outdoor $PM_{2.5}$ (Out_$PM_{2.5}$), residential indoor $PM_{2.5}$ (Ind_$PM_{2.5}$) and workplace $PM_{2.5}$ (Wrk_$PM_{2.5}$) concentrations vs. 48-hour exposure to $PM_{2.5}$ particles from the five source categories identified in sections 5.3.1 and 5.3.2. These results showed that total $PM_{2.5}$ monitoring could predict exposure to inorganic secondary particles (Exp_Seco) better than

Table 4. Model summary of simple linear regressions to predict source attributed 48-hour $PM_{2.5}$ exposure (Exp) for non-ETS participants using personal exposure and microenvironment (residential outdoor (Out), residential indoor (Ind), workplace (Wrk)) $PM_{2.5}$ concentrations as predictors. 'Seco', 'Soil', 'Salt', 'Detr' and 'CoPM' refer to inorganic secondary PM, soil dust, sea salt, detergents and particles from combustion and other sources, respectively.

Dependent	Predictor	n	r^2	Slope	Intercept (µg/m^3)	p-value
Exp_Seco	Exp_$PM_{2.5}$	69	0.62	0.30	0.145	0.000
"	Out_$PM_{2.5}$	69	0.80	0.26	0.251	0.000
"	Ind_$PM_{2.5}$	68	0.58	0.29	0.249	0.000
"	Wrk_$PM_{2.5}$	69	0.30	0.11	1.84	0.000
Exp_Soil	Exp_$PM_{2.5}$	69	0.11	0.10	1.56	0.005
"	Out_$PM_{2.5}$	69	0.02	0.03	2.20	0.285
"	Ind_$PM_{2.5}$	68	0.06	0.07	1.86	0.052
"	Wrk_$PM_{2.5}$	69	0.03	0.03	2.24	0.146
Exp_Salt	Exp_$PM_{2.5}$	69	0.00	0.00	0.214	0.806
"	Out_$PM_{2.5}$	69	0.06	-0.01	0.285	0.040
"	Ind_$PM_{2.5}$	68	0.08	0.00	0.239	0.461
"	Wrk_$PM_{2.5}$	69	0.00	0.00	0.200	0.957
Exp_Detr	Exp_$PM_{2.5}$	65	0.02	-0.01	0.257	0.317
"	Out_$PM_{2.5}$	65	0.05	-0.01	0.276	0.080
"	Ind_$PM_{2.5}$	65	0.00	0.00	0.175	0.832
"	Wrk_$PM_{2.5}$	65	0.04	-0.01	0.265	0.136
Exp_CoPM	Exp_$PM_{2.5}$	65	0.73	0.49	-1.20	0.000
"	Out_$PM_{2.5}$	65	0.35	0.22	0.936	0.000
"	Ind_$PM_{2.5}$	65	0.45	0.32	0.277	0.000
"	Wrk_$PM_{2.5}$	65	0.32	0.26	0.953	0.000

Three outliers removed from analyses with Exp_Detr and Exp_CoPM

exposure to particles from the other four source categories. The models could predict 58 - 80% of the exposure variation, except in workplaces (Wrk_$PM_{2.5}$) where predictions were only 30%. Total $PM_{2.5}$ monitoring could not predict exposures to particles from soil dust, sea salt and detergents. The best of these models could only predict 11% of the variation in exposure to soil particles. Exposure to particles from combustion and other sources (Exp_CoPM) could be moderately predicted (32-45%) by monitoring residential and workplace $PM_{2.5}$ concentrations,

but the best predictor for CoPM was, however, the total $PM_{2.5}$ exposure, explaining 73% of the exposure variation.

A summary of similar regression models using source-attributed concentrations as predictors is presented in Table 5. The two highest explanation rates in these simple linear regression analyses were found in models for predicting exposure to secondary PM. Residential outdoor and indoor concentrations of the secondary PM explained 83% and 81% of the exposure variation. Exposure to soil dust was best predicted by the soil concentration in the workplaces, explaining 47% of the variation. Exposure to sea salt was strongly associated with residential indoor and outdoor concentrations of the sea salt particles explaining 77% and 57% of the exposure variation. Also exposure to detergents could be reasonably predicted by indoor concentrations of detergents. Exposure to CoPM was best predicted by residential indoor concentrations of CoPM, explaining 59% of the exposure variation.

Table 5. Model summary of simple linear regressions to predict source attributed personal 48-hour $PM_{2.5}$ exposures for non-ETS participants using source related personal exposure and microenvironment (residential outdoor (Out), residential indoor (Ind), workplace (Wrk)) $PM_{2.5}$ concentrations as predictors. 'Seco', 'Soil', 'Salt', 'Detr' and 'CoPM' refer to inorganic secondary PM particles, soil dust, sea salt, detergents and particles from combustion and other sources, respectively.

Dependent	Predictor	n	r^2	Slope	Intercept ($\mu g/m^3$)	p-value
Exp_Seco	Out_Seco	66	0.83	0.60	0.123	0.000
"	Ind_Seco	66	0.81	0.71	0.500	0.000
"	Wrk_Seco	68	0.66	0.71	0.627	0.000
Exp_Soil	Out_Soil	66	0.23	0.58	1.67	0.000
"	Ind_Soil	66	0.28	0.52	1.35	0.000
"	Wrk_Soil	68	0.47	0.34	1.56	0.000
Exp_Salt	Out_Salt	66	0.57	0.24	0.113	0.000
"	Ind_Salt	66	0.77	0.41	0.096	0.000
"	Wrk_Salt	68	0.27	0.78	0.088	0.000
Exp_Detr	Out_Detr	N/A	N/A	N/A	N/A	N/A
"	Ind_Detr	63	0.58	0.58	0.090	0.000
"	Wrk_Detr	64	0.06	0.29	0.137	0.063
Exp_CoPM	Out_CoPM	63	0.30	0.34	1.95	0.000
"	Ind_CoPM	63	0.59	0.64	1.33	0.000
"	Wrk_CoPM	64	0.26	0.36	2.38	0.000

N/A = not applicable, three outliers removed from analyses with Exp_Detr and Exp_CoPM

In simple regressions using source related microenvironment predictors, slopes were, as one might expect, higher than in the regressions with $PM_{2.5}$ concentrations as predictors. Slopes ranged from 0.60 to 0.71, 0.34 to 0.58, 0.24 to 0.78, 0.29 to 0.58 and 0.34 to 0.64, in models of exposure to secondary PM, soil dust, sea salt, detergent and CoPM particles, respectively.

Source attributed personal exposures modelled with multiple linear regression using gravimetric microenvironment $PM_{2.5}$ concentrations are summarised in Table 6. The other candidate variables in the step-wise regression analysis are described in section 5.2.1 and in paper III, Table 2b. The highest explanation rate of 83% was for models of Exp_Seco using only Out_$PM_{2.5}$ as a predictor. Exp_Soil was weakly associated with the time of year, explaining 18% of the exposure variation. 21% of the variation in Exp_Salt could be predicted by questionnaire data; the time windows were kept open in homes and the time spent cooking. Models could not predict variation of the Exp_Detr. The best model for Exp_CoPM explained 59% of the exposure variation using Ind_$PM_{2.5}$ and Wrk_$PM_{2.5}$, traffic density of trucks near the home and time spent walking outdoors as predictors.

Source attributed personal exposures were also modelled using source attributed microenvironment concentrations and questionnaire data. Table 7 summarises the models developed in these analyses. Models predicted 79 - 94% of the source attributed exposure variation, except for the model to predict exposure to soil particles, which only predicted 58%.

Table 6. Model summary for multiple stepwise regression of non-ETS exposed participants to predict source related 48-hour $PM_{2.5}$ exposure (Exp). 'Seco', 'Soil', 'Salt', 'Clea' and 'CoPM' refer to inorganic secondary PM, soil dust, sea salt, detergents and particles from combustion and other sources, respectively. The questionnaire variables WINDOW, COOKING, TRUCKSDE and HOMELOCA refer to the time windows were kept open in homes, the time spent cooking, the traffic density of trucks near the home and home location respectively.

Dependent	**Predictors**	**n**	**r^2**	**p-value**
Exp_Seco	Out-$PM_{2.5}$	57	0.83	0.000
Exp_Soil	TIME OF YEAR	57	0.18	0.001
Exp_Salt	WINDOW, COOKING	57	0.22	0.001
Exp_Detr	TRUCKSDE	55	0.08	0.035
Exp_CoPM	Ind_$PM_{2.5}$, TRUCKSDE, HOMELOCA	55	0.59	0.000

Three outliers removed from analyses with Exp_Detr and Exp_CoPM

Finally, Table 8 summarises multiple regression models to predict source attributed personal exposures, in the absence of measured indoor microenvironment data (compare to III; Table 2c). Again, exposure to secondary PM could be predicted with great detail, exposures to CoPM and sea salt could be modelled fairly well. The weakest predictors of exposures were found for soil particles.

Table 7. Model summary for multiple stepwise regression of non-ETS exposed participants to predict source related 48-hour $PM_{2.5}$ exposure (Exp). Microenvironment $PM_{2.5}$ concentrations replaced by source attributed $PM_{2.5}$ concentrations in each microenvironment. 'Seco', 'Soil', 'Salt', 'Detr' and 'CoPM' refer to inorganic secondary PM, soil dust, sea salt, detergents and particles from combustion and other sources, respectively. The new questionnaire variables (additional to Table 6) BUS and STOVE refer to the time spent in bus and the type of stove in residence, respectively.

Dependent	Predictors	n	r^2	p-value
Exp_Seco	Out_Seco, Wrk_Seco, Ind_Seco, BUS	54	0.94	0.000
Exp_Soil	Wrk_Soil, Out_Soil	54	0.58	0.000
Exp_Salt	Ind_Salt, Wrk_Salt, WALK	54	0.82	0.000
Exp_Detr	Ind_Detr, Wrk_Detr	52	0.80	0.000
Exp_CoPM	Ind_CoPM, Wrk_CoPM, HOMELOCA, STOVE, TRUCKS	52	0.79	0.000

Three outliers removed from analyses with Exp_Detr and Exp_CoPM

Table 8. Model summary for multiple stepwise regression of non-ETS exposed participants to predict source related 48-hour $PM_{2.5}$ exposure (Exp). Source related residential indoor and workplace concentrations excluded from input variables. A new questionnaire variable (additional to Table 6 and Table 7) TRAIN refers to the time spent in train.

Dependent	Predictors	n	r^2	p-value
Exp_Seco	Out_Seco, WINDOW	55	0.85	0.000
Exp_Soil	Out_Soil	55	0.25	0.000
Exp_Salt	Out_Salt, TRAIN	55	0.55	0.000
Exp_CoPM	Out_CoPM, HOMELOCA	53	0.53	0.000

Three outliers removed from analysis with Exp_CoPM

6 DISCUSSION

6.1 $PM_{2.5}$ exposure studies

Historically most research groups have had to develop their own monitors and other equipment needed in the exposure studies, due to lack of commercial standard equipment and monitors for personal exposure monitoring. This has a lead to a variety of monitors and equipment used. Because of this variety of monitors, it is important to evaluate the precision of the methods before the results of these studies can be compared. Quality assurance results of large-scale exposure studies are discussed in the following paragraphs.

A small $PM_{2.5}$ inertial impactor (Marple Scientific Products, Inc.) was constructed for use with pumps at 2 L/min for the Toronto study (Pellizzari et al. 1999). This monitor was remarkably lightweight (820 g) and it could monitor for the 3-day sampling period with only one set of batteries using a duty cycle of 0.75 (3 min on, 1 min off). This PEM was carried in a waist pack. Overall precision (relative standard deviation, RSD) for co-located monitors ranged from 2.5% to 5.0% agreeing with the RSDs of 2.6% for PEM and 3.1% for MEM in the current study. The limits of detection (LOD) in the current study for PEM, 2.8 $\mu g/m^3$, and for MEM, 1.0 $\mu g/m^3$, were lower than the LOD of the PEM in the Toronto study, 3.5 $\mu g/m^3$.

In the tobacco study reported by Phillips et al. (1996, 1998a, 1998b, 1999) PEM consisted of a 10 mm Dorr-Oliver cyclone, designed for monitoring RSP ($PM_{3.5}$) exposures at flow rate of 1.72 L/min, a battery-operated pump and a polypropylene box. The sampling head was attached to the shoulder strap in the breathing zone of the study subject for the 24-hour sampling period. Using this method considerably high LODs, 8.2 $\mu g/m^3$, were reported for 24-hour sampling period compared to the LODs of *EXPOLIS* and Toronto studies. In addition to these two large-scale exposure studies, quality assurance results of the current study are compared to those of other exposure studies in paper I.

In general personal exposures to $PM_{2.5}$ for all the studied participants were lower in Helsinki compared to all other *EXPOLIS* centres (Jantunen et al. 1999). Mean personal daytime exposures in Helsinki, Basel, Prague, and Athens were 19 $\mu g/m^3$, 24 $\mu g/m^3$, 47 $\mu g/m^3$ and 72 $\mu g/m^3$ respectively. Similar patterns were found for personal night time exposures and consequently for 48-hour exposures to $PM_{2.5}$.

Personal exposures to $PM_{2.5}$ in a population of adults in Toronto (Pellizzari et al. 1999) and exposure to RSP (PM_3) among children in the Netherlands (Janssen et al. 1999) were about twofold (28.4 µg/m^3 and 28.3 µg/m^3 respectively) compared to Helsinki (see Figure 3). The most important factor low exposures in Helsinki is due to low regional background levels (Ojanen et al. 1998). The number of cars in Helsinki is lower than in Prague and Athens, therefore fine particle emissions from traffic may also be lower. Similarly, local emissions from heating are low, because almost all houses are heated by a district heating system of hot water pipelines from large co-generating power plants. In addition, Helsinki is situated next to the sea, and local emissions are often diluted with relatively clean marine winds. In addition, wood burning and gas cooking are rare. Consequently, residential indoor concentrations in Helsinki were also about half those in Toronto and Basel, and about one third those in Athens, Milan and Prague. Lower residential indoor concentrations in Helsinki may also be the result of a small number of residences where people smoke indoors.

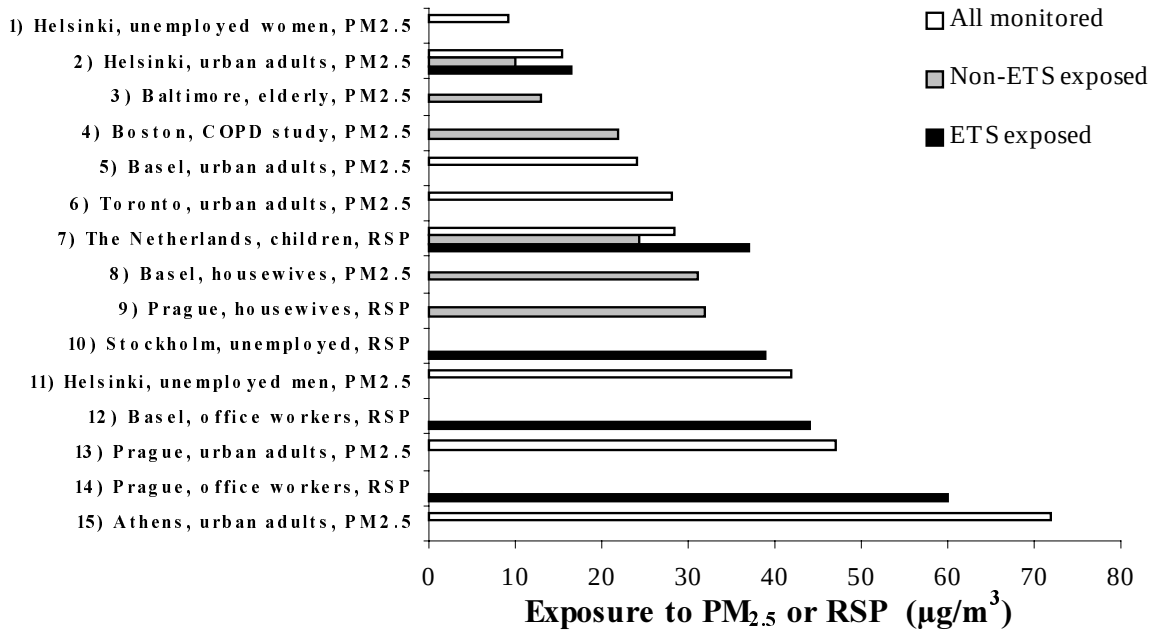

Figure 3. Summary of the results reported in the exposure studies. Numbers 1)-15) refer to the following sources: 1), 2) and 11) this thesis, 3) Williams et al. 2000, 4) Rojas-Bracho et al. 2000, 5) , 13) and 14) Jantunen et al. 1999, 6) Pellizzari et al. 1999, and 7) Janssen et al. 1999. Phillips and his co-authors have reported results from studies 8) and 12) (1999), 9) and 14) (1998b), and 10) (1996).

Mean 48-hour population exposures to $PM_{2.5}$ in the current study (Table 3) were about one half of the lowest mean 24-hour population exposure to RSP found by Phillips and his co-authors for workers and housewives in Stockholm, Bremen, Prague and Basel (Phillips et al. 1996, Phillips et al. 1998a, Phillips et al. 1998b, Phillips et al. 1999). It must be noted here that slightly higher concentrations are expected because of slightly higher cut-size of the RSP compared to $PM_{2.5}$. Similar exposure levels were found among the population of non-working men and women living in homes in Stockholm with residents that smoked by Phillips et al. (1996), compared to the population of unemployed men in *EXPOLIS*-Helsinki. In Bremen the median 24-hour exposure to RSP in the population of workers living and working with smokers was about 36 times (789 $\mu g/m^3$ vs. 22 $\mu g/m^3$, values not shown in Figure 3) the highest median exposure to $PM_{2.5}$ found in any sub-population of the current study (unemployed men) in Helsinki. The mean exposure to RSP found in the population of the housewives living in non-smoking households in Prague (Phillips et al. 1999) and Basel (Phillips et al. 1998b) were threefold compared to the population of the unemployed women in Helsinki.

Mean personal 48-hour $PM_{2.5}$ exposures of non-ETS exposed participants in the current study were about half the 12-hour $PM_{2.5}$ exposures of individuals with COPD disease in Boston (Rojas-Bracho et al. 2000). Similarly, mean residential indoor concentrations were about two times higher in the COPD study and mean ambient $PM_{2.5}$ concentration 1.5 times higher compared to the current study.

Mean personal exposures to $PM_{2.5}$ in the non-smoking population of elderly people living at a retirement centre in Baltimore, USA (Williams et al. 2000) were slightly higher compared to the sub-population of adults not exposed to ETS in the current study. Residential indoor $PM_{2.5}$ concentrations were similar for the sub-population of non-ETS exposed adults in the current study and the elderly in the Baltimore study. Mean ambient $PM_{2.5}$ concentrations in Baltimore were about two times concentrations in Helsinki. Similar mean personal exposure levels in the current study and in the Baltimore study reflect indoor concentrations and, thus suggest that there were few or no major indoor sources, in addition to smoking, in these living environments. Interestingly, similar indoor $PM_{2.5}$ concentrations were found even though ambient $PM_{2.5}$ concentrations were twofold in Baltimore. This result suggests that building shell can reduce indoor exposure to $PM_{2.5}$ from ambient origin.

It must be noted that there are neither European nor Finnish national air quality guidelines for $PM_{2.5}$ when comparing our results from *EXPOLIS*-Helsinki to current air quality regulations. In the US a new National Air Quality Standard (NAAQS) was promulgated in 1997 for particulate matter including guidelines for ambient $PM_{2.5}$ concentrations (EPA 1997). The annual average $PM_{2.5}$ guideline value was set to 15 $\mu g/m^3$ and 24-hour average to 65 $\mu g/m^3$. In the current study the mean ambient $PM_{2.5}$ concentration in Helsinki, which can be compared to annual average values as the study period was approximately one year, did not exceed this guideline value. Mean population exposures to $PM_{2.5}$, however, were clearly higher than the mean ambient concentration and were similar to the annual guideline value. Recent health effect studies have shown that $PM_{2.5}$ can cause adverse health effects at annual mean concentration levels below 20 $\mu g/m^3$ and the available information does not indicate a threshold level below which no effects would be expected (WHO 2000). Therefore we can assume that even these considerably low $PM_{2.5}$ exposure levels found in this study may have adverse health effects on the population of Helsinki.

6.2 Determinants of the $PM_{2.5}$ exposures

6.2.1 Behavioural and microenvironmental determinants

Smoking was found to be by far the most important determinant for personal exposure to $PM_{2.5}$ in the current study. Similarly, smoking has been clearly the strongest determinant of the fine particles for personal exposure and indoor concentration in several other studies (Pellizzari et al. 1999, Özkaynak et al. 1996, Wallace 1996). In addition to smoking, outdoor air is the other major source of particles in indoor air and subsequently in personal exposures of participants of *EXPOLIS*-Helsinki. This result agrees well with results reported by the Dutch and in the Toronto and in the PTEAM studies (Janssen et al. 1999, Pellizzari et al. 1999, Özkaynak et al. 1996). The association between personal exposure and residential indoor $PM_{2.5}$ concentration was strong and very similar in Toronto and *EXPOLIS*–Helsinki studies showing that personal exposures are closely related to indoor concentrations. The associations between personal exposure and residential outdoor and fixed site $PM_{2.5}$ concentrations, however, were weak in both of these studies. In the Dutch study (Janssen et al. 1999), the correlation between exposure and ambient fine particle concentration also was analysed using longitudinal data on the same individuals. Such correlation was, as one might expect, higher than the cross sectional correlation observed in the current and the Toronto study. The correlation between exposure

and ambient fine particle concentration increases, when repeated measurements are analysed (longitudinal analysis) instead of pooling from exposures of individuals who were measured only once (cross-sectional analysis).

Residential and workplace $PM_{2.5}$ concentrations, traffic density near home and home location were significant determinants for $PM_{2.5}$ exposure of participants in the current study using multiple regression analysis. This result confirms the results of the simple linear regression analyses showing the strong association between residential and workplace indoor concentrations and exposure. Traffic density near home and home location, both reflect the presence of the link between increased traffic density and the increased $PM_{2.5}$ exposure. 'Season', 'keeping windows open in homes' and 'home situated downtown' were also significant determinants for exposure to $PM_{2.5}$ when analysing sub-sets of data. Air exchange rate, which is similar to time windows were open in the current study, was also found as a determinant for residential indoor $PM_{2.5}$ concentration in the PTEAM study (Özkaynak et al. 1996). It must be noted that the temporal variation in ambient background concentrations may cause bias to the comparisons between sub-populations. If the number of observations is large enough (tens of cases), the cases are assumed to be randomly distributed to studied categories eliminating the possible bias. With low number of cases in a category, probability of the biased results may be increased.

6.2.2 Socio-demographic determinants

Analysis of socio-demographic variables identified occupational status, level of education and age as the best descriptors for exposure to $PM_{2.5}$. Those with a lower occupational status, less than 14 years of education and 35 years of age had higher exposures than those with higher occupational status, more than 14 years of education and 35 years of age. Workplace concentrations and smoking explained most but not all of the socio-economic differences. Personal day and night time exposures and $PM_{2.5}$ concentrations in the homes resulted in $PM_{2.5}$ exposure differences between age groups. Men had on average higher exposures and larger exposure differences between the socio-demographic groups than women. Interestingly, differences were not observed between genders, different socio-economic groups or age groups in residential outdoor $PM_{2.5}$ concentrations. Exposure to environmental tobacco smoke did not create new differences between the socio-demographic groups; instead, it amplified the pre-

existing differences. These results agree with the results published by Cavelaars et al. (1998) showing that smoking prevalence was higher in lower socio-economic status populations.

These results show that significant differences exist between different socio-demographic sub-populations. Yet, socio-demographic determinants alone were poor predictors of population exposures to $PM_{2.5}$ in multiple regression analysis. Socio-demographic determinants are poor predictors of exposure in regression analyses, because they are not causal, instead they indicate relative differences in behavioural and microenvironment determinants such as housing, smoking, and higher workplace exposures. Socio-demographic descriptors can, however, improve the population exposure models based on environmental variables. This association was demonstrated in multiple regression analysis to predict population exposure to $PM_{2.5}$ without residential and workplace indoor $PM_{2.5}$ concentrations, where residential outdoor $PM_{2.5}$ concentrations, 'years of education', 'age' and 'traffic density on the street nearest the home', explained the exposure variation better than models without socio-demographic variables (55% and 47% respectively).

6.3 Source apportionment

In agreement with determinant analysis smoking was identified as the strongest source for $PM_{2.5}$. As a source linked to human behaviour, it especially increased exposures of active smokers. It was also the strongest factor that increased indoor $PM_{2.5}$ concentrations if present, and on average was directly responsible for the largest proportion of exposure to $PM_{2.5}$ for non-smokers that live or work in the presence of smokers. Not surprisingly, smoking has been reported to be the major determinant of exposure to fine particles in several other studies including the European tobacco study (Phillips et al. 1996, 1998a, 1998b, 1999) and in the Toronto study (Pellizzari et al. 1999).

Figure 4 shows the slightly different source categories used in the PCA and in the source reconstruction (SR) for clarification. It must be noted that the category 'CoPM' in SR consist of both, local and long-range transported combustion-originated particles. Similarly, the category 'long-range transported particles' (LRT) in PCA is a combination of secondary and primary PM.

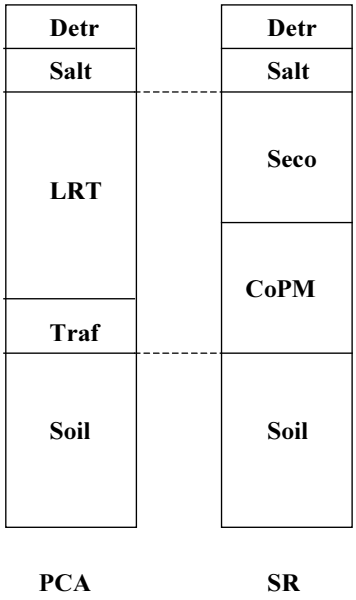

Figure 4. The relationship between the source categories identified in the PCA and in the source reconstruction (SR). In the PCA following factors were identified: soil PM (soil), local combustion, mostly traffic (Traf), long-range transported PM (LRT), sea salt (Salt) and detergent PM (Detr). In SR different categories are 'combustion and other particulate matter' (CoPM) and inorganic secondary PM (Seco).

PCA and source reconstruction (SR) were applied to identify and quantify other sources than smoking. In residential outdoor air inorganic secondary PM and particles from 'combustion and other sources' (CoPM) were the major contributors of the total $PM_{2.5}$ concentration (V; Figure 1). Inorganic secondary PM is mostly the result of long-range transport from precursor sources such as traffic, industrial combustion and power stations emitting NO, burning of sulphur containing fuels for SO_2 and animal wastes and industrial fertilisers producing NH_3 emissions. For example photochemical oxidation of SO_2 under typical ambient conditions occurs on a time-scale of 30-50 hours (APEG 1999). Local precursor sources, therefore, usually contribute minimally to sulphate $PM_{2.5}$ concentrations in urban areas because of high stacks and prevalent winds. The composition of ambient fine particles in the current study agreed with the results reported by Ojanen et al. (1998) and Pakkanen et al. (2001a), also from the Helsinki area. Similar ambient air fine particle compositions in California were reported by Schauer et al (1996).

The same four source categories, secondary PM, CoPM, soil dust and sea salt PM were found in residential indoor samples and outdoor samples. Residential indoor concentrations for the particles of ambient origin, i.e. secondary PM, CoPM and sea salt PM were decreased by about

one third compared to their residential outdoor concentrations. Thus, residential building shells seem to restrict the infiltration of particles from outdoor sources. This finding agrees with the conclusions drawn by Sarnat et al. (2000) and Long et al. (2001). In contrast, mean concentrations of soil particles in residential indoor air were 56% higher compared to outdoor air showing a strong indoor source for soil particles. Such an indoor source has obviously arisen through the re-suspension of tracked in dust from indoor surfaces. This finding was also confirmed by I/O ratios equal or exceeding 1 in all seasons for elements typically associated with soil minerals (V; Table 6). Similarly, Yakovleva et al. (1999) identified three categories 'ambient soil', 'indoor soil', and 're-suspended indoor soil' from the PTEAM data from Riverside, CA, USA. They identified these soil categories using PM_{10} data, but even though soil dust is mainly present in coarse particle fraction ($PM_{2.5-10}$), a lesser contribution is still present in fine fraction ($PM_{2.5}$). The composition of the indoor $PM_{2.5}$ identified in the current study in Helsinki agrees with the sources, 'personal activities', 'secondary sulphate', 'motor vehicle exhaust' and 'soil', reported by Yakovleva et al. (1999) in Riverside, CA, USA.

In addition to particles of ambient origin one indoor source, detergent PM, was identified only from residential indoor air. Interestingly, the detergent factor in PCA (V; Table 5) had quite a strong association with the total elemental variation (14%) compared to its low contribution to the total indoor $PM_{2.5}$ concentration. The fact that there were a few samples (present in exposures and in indoor air) with quite high phosphorus concentrations, obviously from similar conditions, in the elemental data, may explain this discrepancy.

Although, the I/O ratio for the total $PM_{2.5}$ mass concentrations was close to unity, the I/O ratios of the elements varied considerably depending on the season and could be grouped according to source compositions (V; Table 6). This result shows that even if the I/O ratio for $PM_{2.5}$ is close to unity, the composition of the aerosol changes significantly from ambient to indoor air. This finding also suggests that particles from different outdoor sources do not penetrate equally into indoor environments. The obvious reason for these differences was that infiltration depends on particle size, which was demonstrated by Long et al. (2001).

I/O ratios for the elements that were associated with long-range transport were approximately 0.6-0.7 in winter and spring and approached 1 in summer. The increased infiltration of these elements in summer could be explained by strongly increased open window ventilation. A slightly different pattern of I/O ratios was observed for elements that were associated with

CoPM. These elements showed remarkably similar I/O ratios in spring, summer and fall but much decreased penetration in the winter. According to Götschi et al. (2002), in addition to the lower concentrations of $PM_{2.5}$ and black smoke in Helsinki, mean I/O ratios were also lower than those in other *EXPOLIS* centres Athens, Basel and Prague. Comparison of the black smoke showed that there were also less indoor sources for black smoke or buildings were tighter in Helsinki than in other centres. This is credible, because the levels of indoor smoking, gas cooking and the frequency of indoor combustion devices are all lower in Helsinki than in the other *EXPOLIS*–cities. Similarly, winter/summer ratios for both $PM_{2.5}$ and black smoke in ambient air were lowest in Helsinki presumably due to the virtual absence of space heating emissions during the winter; practically all buildings are heated by district heating pipelines.

The composition of $PM_{2.5}$ in workplaces was very similar to the composition in residential indoor air. Small differences were found only for the mean CoPM and for detergent contributions. Higher mean CoPM contribution in workplaces may reflect the presence of occupational sources or stronger contribution of traffic emissions in daytime. Lower mean detergent contribution is due to fewer cleaning activities in workplaces that use detergent compared to in homes.

Finally, the composition of exposure samples reflected the different microenvironments in which participants spent their time. Exposures to secondary particles and sea salt were about two thirds of their outdoor air concentrations. This difference reflects typical infiltration rates 0.65 – 0.70 (Wallace 1996) for ambient fine particles, reducing indoor exposures as people spend most of their time indoors and few or no indoor sources are expected for these particles. Indoor concentrations, therefore, strongly drive exposures. Mean exposures to CoPM were only slightly lower than mean CoPM concentrations in residential outdoor air, but higher than residential indoor concentrations. These results suggest that individuals have been exposed to higher CoPM concentrations than one might expect based on the outdoor and indoor concentrations probably due to the time spent in the vicinity of traffic (commuting) and indoor cooking. Personal exposures to soil dust were higher than residential outdoor concentrations, but similar to indoor concentrations, reflecting the strong contribution of indoor sources to personal exposures. Thus surprisingly, people are exposed mostly to soil particles in indoor environments, although personal exposures to soil dust may consist of two components, one reflecting re-suspension and indoor activities and the other reflecting variation of soil dust in outdoor air. In this study, a few exceptionally high phosphorus concentrations were detected in

residential indoor air and in corresponding exposure samples. Therefore, exposure to detergents is clearly associated with certain cleaning activities or products used in residences.

In the source reconstruction method category CoPM includes PM emissions from traffic and cooking among others. It would have been better if for example traffic and cooking could have been analysed separately, but it was not possible in this study because valid trace elements were not available for those categories. The category CoPM will be separated to smaller categories in the forthcoming studies if valid methods can be found.

6.4 Applicability of ambient and indoor monitoring for personal exposure assessment to $PM_{2.5}$ from different sources

Personal exposures to inorganic secondary $PM_{2.5}$ were better predicted by microenvironment $PM_{2.5}$ monitoring concentrations than personal exposures to particles from any other source category. Using single microenvironment concentrations as predictors, personal exposures to secondary PM could be equally (r^2 = 0.8) predicted by either residential outdoor $PM_{2.5}$ or secondary PM concentrations from either residential outdoor or indoor air. Questionnaire variables did not remarkably improve predictive ability of the models, when used as candidate variables in multiple stepwise regressions. Instead, the highest predictive ability of models was found, when secondary PM concentrations in residential outdoor, indoor air and in workplaces were used together with the questionnaire variable ‘time spent in a bus’. This is not surprising as secondary PM has only ambient sources and is one of the major contributors to the total $PM_{2.5}$ mass concentration. Oglesby et al. (2000a) also found a high correlation (r^2 = 0.72) between exposures and ambient sulphur concentrations in Basel, Switzerland, which agreed well with that of the current study (r^2 = 0.83). In addition, Wallace (1996) presented pilot results of the PTEAM study from California showing very similar associations between exposure and ambient daytime $PM_{2.5}$ sulphur (r^2 = 0.78) compared to respective associations for secondary PM in the current study.

Exposure to $PM_{2.5}$ was clearly a better predictor of exposure to CoPM than residential indoor $PM_{2.5}$ concentration. This result suggests that CoPM has, in addition to ambient and indoor sources also significant behavioural sources. Such sources might be for example traffic and indoor cooking. The role of traffic in increased personal exposures to CoPM was confirmed by better prediction of exposure to CoPM using residential outdoor $PM_{2.5}$ concentrations together

with home location, compared to residential outdoor $PM_{2.5}$ concentrations alone. Similarly, traffic density on streets next to residences, home location and residential indoor $PM_{2.5}$ concentrations were better predictors of exposure to CoPM than residential indoor $PM_{2.5}$ alone. Residential and workplace indoor CoPM concentrations combined with home location, type of stove and traffic density on the streets next to residences were the best predictors of personal exposures to CoPM. This result suggests cooking as a contributor to CoPM exposures.

In the Toronto study (Pellizzari et al. 1999) variation of exposure to $PM_{2.5}$ manganese (Mn) was predicted by residential indoor and outdoor $PM_{2.5}$ Mn concentrations explaining 31%, and 24% respectively. A manganese based compound (MMT) has been used as a gasoline additive in Canada for about 20 years and therefore these results can be best compared to those of CoPM in the current study. Association between exposure variation and residential indoor and outdoor concentrations of CoPM were higher in *EXPOLIS*-Helsinki explaining 59% and 30% of the exposure respectively. Similar associations between ambient and exposure concentrations of $PM_{2.5}$ bound lead ($r^2 = 0.28$) in Basel, which were used as a tracer of traffic, were reported by Oglesby et al. (2000a). Correlations between personal exposure to $PM_{2.5}$ and residential indoor $PM_{2.5}$ concentrations in the Toronto study showed slightly higher association ($r^2 = 0.62$) compared to the current study ($r^2 = 0.53$). Ambient $PM_{2.5}$ concentrations were also poor predictors of personal exposures in the Toronto study.

Exposure to soil PM could not be predicted using total $PM_{2.5}$ monitoring probably due to several source types of soil PM. Exposure to soil was best predicted by the time of the year alone, even though the association was weak ($r^2 = 0.18$), when using total $PM_{2.5}$ concentrations together with questionnaire variables in multiple regression analysis. This is not surprising as the Finnish climate has clearly defined seasons with considerable time during the year when the ground is covered with snow and ice, which limits re-suspension of soil particles. Interestingly, soil PM concentrations in the workplace were more strongly associated with exposures to soil PM than residential soil PM concentrations. Mean residential indoor and workplace soil PM concentrations, however, were very similar. Clearly, higher associations between workplace concentrations and exposures to soil PM were due to large variation of soil PM concentrations in workplaces.

Sea salt and detergent PM could not be predicted using total $PM_{2.5}$ monitoring probably due to the low contribution of these source categories to total $PM_{2.5}$ concentrations. Residential indoor and workplace concentrations of these particles, however, were good predictors of exposure to detergents and, when combined with time spent walking outdoors, were good predictors (r^2 = 0.8) of exposure to sea salt. These results suggest that exposure to sea salt and detergent PM can be predicted using microenvironment modelling with source attributed concentrations.

The y-axis intercept of linear regression of residential indoor $PM_{2.5}$ concentrations to predict personal exposure can be used as an estimate of 'personal cloud' (see Table 5). Similar personal cloud effects of 1.3 $\mu g/m^3$ were suggested for both CoPM and soil PM. In the absence of smoking and fireplaces it is plausible that people's exposure to CoPM was higher than residential indoor concentrations due to commuting and being outdoors in the proximity of traffic. Exposure to soil PM was caused by re-suspension, and thus human activities, and therefore could be higher than the respective residential indoor concentration. Personal cloud effects of detergent $PM_{2.5}$ sources were weak.

Ambient concentrations of $PM_{2.5}$ have been linked to adverse health effects (Dockery et al. 1993), but the mechanisms and constituents of $PM_{2.5}$ causing these effects are not known. After health relevant $PM_{2.5}$ constituents have been identified, cost- effective actions can be carried out to reduce these health effects, if exposure to these particles is known. This study has provided differences in exposure to $PM_{2.5}$ originated from the main sources. These results showed high association between ambient total $PM_{2.5}$ concentrations and exposure to ambient originated inorganic secondary particles, but no association with soil PM. Thus, the results of this study may be used in the future to evaluate the validity of ambient air monitoring to assess exposure to harmful $PM_{2.5}$ constituents.

7 CONCLUSIONS

The precision of microenvironment (MEM) monitors was better than the precision of personal (PEM) monitors developed for this study, and both were equal to or better than in those used in previous exposure studies. In addition, comparative tests showed good agreement between these two monitoring methods. Analysis of sources of error in differential weighing of blank filters indicated that one third of the total error, after statistic charge removal, was correctable. The most important sources of correctable error were air buoyancy variation and filter storage time.

Personal 48-hour exposures to $PM_{2.5}$ in Helsinki were low compared to those that have been measured in most other studies in North America and Europe. The cross sectional distribution of 48-hour exposures across individuals and time was log-normal, with arithmetic mean of 15.4 $\mu g/m^3$, geometric mean of 10.9 $\mu g/m^3$, and geometric standard deviation of 2.13 $\mu g/m^3$ for the whole study population. Average workday exposure levels were 48% higher than average leisure time exposure levels. Currently, there are no air quality guidelines for $PM_{2.5}$ in Finland, but population exposure levels in Helsinki did not exceed ambient air quality standards for annual average $PM_{2.5}$ concentrations in the USA.

The most important determinant of personal exposures to $PM_{2.5}$ was presence of tobacco smoking. Unless stated otherwise, henceforward all conclusions concern individuals who did not smoke and did not report exposure to environmental tobacco smoke (ETS). Residential outdoor and ambient fixed site concentrations did not adequately predict personal exposures, and personal exposures were usually higher than $PM_{2.5}$ concentrations in these microenvironments. Residential and workplace indoor concentrations combined with the traffic density on the streets next to residences were the best determinants of exposures. Probably reflecting greater traffic density, personal exposures downtown were on average one third higher than exposures in the suburbs.

Smoking, occupational status, education and age were the best determinants to predict differences in $PM_{2.5}$ exposures between socio-demographic sub-populations. Lower occupational status, low education, and age less than 35 years reflected higher exposure compared to upper status, higher education and higher age. The presence of ETS amplified exposure differences between socio-demographic sub-populations, but did not fully explain

them. Men had higher exposures and larger exposure differences between the socio-demographic sub-populations than women partly as a result of exposure to ETS.

Major source categories of $PM_{2.5}$ personal exposures identified in this study were smoking, inorganic soil components, primary and secondary long-range transported particles, and local combustion sources mostly consisting of traffic. Inorganic secondary particles were mostly from sources located some distance from Helsinki and were transported into Helsinki. Primary particles originated both from local sources in the immediate vicinity and long distance sources. Major sources of soil dust were in the residential indoor environment and indoor-originated soil dust was responsible of more than half the personal exposures to soil particles. Thus, ambient monitoring of $PM_{2.5}$ under-estimated the contribution of soil particles to personal exposures. In contrast, ambient monitoring over-estimated personal exposures to particles from outdoor sources such as inorganic secondary particles by approximately two thirds.

Ambient total $PM_{2.5}$ mass variation was a moderate predictor of $PM_{2.5}$ exposure variation within the population (regression $r^2 = 0.4$). When total $PM_{2.5}$ mass was apportioned into contributions from each of the main sources, ambient monitoring of inorganic secondary PM was a good ($r^2 = 0.8$) predictor of inorganic secondary PM exposures. Similarly ambient monitoring of CoPM was a moderate predictor ($r^2 = 0.3$), and ambient monitoring of soil PM a weak predictor ($r^2 = 0.2$) of respective exposures. As expected, total ambient $PM_{2.5}$ mass was generally a weaker predictor for exposure to PM from each of these 3 sources.

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