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$PM_{2.5}$ AND NO_2 EXPOSURE ASSESSMENT OF URBAN POPULATION IN THE HELSINKI METROPOLITAN AREA AND OTHER EUROPEAN URBAN AREAS

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Academic dissertation

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$PM_{2.5}$ and NO_2 exposure assessment of urban population in the Helsinki Metropolitan Area and other European urban areas

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University of Helsinki, 2002

Abstract

Epidemiological literature of the 1990s revealed surprisingly large public health impacts associated with the present common levels of ambient PM_{10} and $PM_{2.5}$ (particulate matter with an aerodynamic diameter of less than 10 µm and 2.5 µm, respectively) in European as well as North American cities. In addition, these health effects are apparently more serious than earlier assumed. Harmful health effects and exposure to NO_2 (nitrogen dioxide) have also been associated with concentrations below the ambient air quality guideline levels set by the World Health Organization.

City air quality is monitored by ambient fixed site monitoring networks and exposures to air pollutants are assessed by these monitors. In addition to ambient air quality, researchers have started to discuss the roles of behavioural (personal) and indoor sources in human exposure. It is not clear how well ambient fixed site concentrations represent total personal exposures of populations or differences between populations.

This thesis introduces different ways to evaluate $PM_{2.5}$ and NO_2 exposures in urban areas. In the EXPOLIS study (air pollution exposure distributions within adult urban populations in Europe), total personal exposures and microenvironment concentrations of $PM_{2.5}$ and NO_2 were measured in the most important microenvironments (homes and workplaces). Using this data, associations and dependencies between total personal exposures, microenvironment concentrations and ambient fixed site monitoring results were studied. In the EXPAND project (EXPosure to Air pollution, especially to Nitrogen Dioxide and particulate matter), a model for evaluating the exposure of an urban population to ambient air pollution was developed and the results were illustrated using the Geographic Information System. The results of this model were compared with measured results of EXPOLIS.

The study revealed that although ambient fixed site $PM_{2.5}$ concentrations were evenly distributed in the Helsinki Metropolitan Area the concentrations measured at ambient fixed site could explain only 15% of the total personal (cross-sectional, short-term) exposure variation. Total exposures during non-working hours correlated better than those of working hours with outdoor/ambient concentrations. Therefore, $PM_{2.5}$ data from the ambient fixed site monitors can adequately explain the total short-term $PM_{2.5}$ exposure of less active population, such as retirees, but not that of the active working age population. It was further shown that ambient fixed site NO_2 explained only 19% of the total personal (cross-sectional, short-term) exposure variation when data from Helsinki, Basel and Prague were combined (11% Helsinki data only). It should be noted, though, that the use of personal questionnaire information can significantly improve the predictive power of NO_2 concentrations measured at ambient fixed sites or the residential outdoors. When compared with measured data, the developed exposure model is quite promising

Keywords: exposure assessment, $PM_{2.5}$, NO_2, exposure modelling

Contents

List of original publications

This thesis is based on five original publications, referred to in the text by their Roman numerals (I-V), and additional analyses completed for this thesis. Papers are reproduced with the kind permission of the journals concerned.

I Pohjola, M., Kousa, A., Kukkonen, J., Härkönen, J., Karppinen, A., Aarnio, P., Koskentalo, T., 2002. The spatial and temporal variation of measured urban PM_{10} and $PM_{2.5}$ in the Helsinki Metropolitan Area. Water, Air & Soil Pollution: Focus (in press).

II Kousa, A., Oglesby, L., Koistinen, K., Künzli, N., Jantunen, M., 2002. Exposure chain of Urban Air $PM_{2.5}$ - Associations between ambient fixed site, residential indoor, outdoor, workplace and personal exposures in the European EXPOLIS study. Atmospheric Environment, 36, 3031-3039.

III Kousa, A., Monn, C., Rotko, T., Alm S., Oglesby L. Jantunen, M., 2001. Personal exposures to NO_2 in the *EXPOLIS*-study: Relation to residential indoor, outdoor and workplace concentrations in Basel, Helsinki and Prague. Atmospheric Environment, 35, 3405-3412.

IV Kousa, A., Kukkonen, J., Karppinen, A., Aarnio, P., Koskentalo, T., 2001. Statistical and diagnostic evaluation of a new-generation urban dispersion modelling system against an extensive dataset in the Helsinki area. Atmospheric Environment, 35, 4617-4628.

V Kousa, A., Karppinen, A., Aarnio, P., Koskentalo, T., Kukkonen, J., 2002. A model for evaluating the exposure of an urban population to ambient air pollution. Atmospheric Environment, 36, 2109-2119.

1 Introduction

Atmospheric aerosol particles have a considerable effect on our lives. Airborne particulate matter represents a complex mixture of organic and inorganic substances, their composition being mainly dictated by their formation processes. Particles which are more than 1 μm in diameter are formed by mechanical processes, and particles less than 1 μm in diameter are formed by gas-to-particle conversion. The recent development of more sensitive instrumentation has enabled observations of new particle formation events. Observations have been made in locations ranging from rural and remote sites to urban environments (Raes et al., 1997; O'Dowd et al., 1999; Kulmala et al., 2000a; Väkevä et al., 2000; Aalto et al., 2001). The events have also been predicted through theoretical models (e.g. Kulmala et al., 1998, 2000b). A fraction of these new particles survive to grow into larger particles through condensation and coagulation processes. Because of increased anthropogenic emissions, the number and mass of atmospheric aerosols have increased over the past century. Atmospheric particles have been implicated in the reduction of visibility in urban and regional areas, in acid deposition and in perturbing the earth's radiation balance (Seinfield and Pandis, 1998). Particles affect the earth's radiative balance directly, by scattering light, and indirectly, by acting as cloud condensation nuclei, thereby influencing the albedo and life-time of clouds.

Atmospheric aerosols also affect our health. Epidemiological literature of the 1990s revealed surprisingly large public health impacts associated with present levels of ambient PM_{10} and $PM_{2.5}$ (particulate matter with an aerodynamic diameter of less than 10 μm and 2.5 μm, respectively) in European as well as North American cities. Futher, these health effects have turned out to be more serious (Dockery et al., 1993; Schwartz et al., 1996; Katsouyanni et al., 1997; Pekkanen et al., 1997; Künzli et al., 2000; Penttinen et al., 2001; Pope et al., 2002) than earlier assumed (e.g. WHO, 1987). Harmful health effects and exposure to nitrogen dioxide (NO_2) have also been associated with concentrations below ambient air quality guideline levels set by the World Health Organization (WHO) (Brunekreef et al., 1989; Schwartz et al., 1991; Mukala et al., 2000).

City air quality is monitored by ambient fixed site monitoring networks and exposures to air pollutants are assessed by these monitors. In addition to ambient air quality, researchers have started to discuss the roles of behavioural (personal) and indoor sources in human exposure. It is not clear, how well ambient fixed site concentrations represent total personal exposures of populations or differences between populations. In developed countries, people spend most of their time in indoor environments (e.g. 87% in Helsinki, based on EXPOLIS time-activity data) such as homes and

workplaces. Therefore, it is important to measure the total personal exposures and also the microenvironment concentrations in the relevant environments. This allows associations and comparisons to be drawn between total personal exposure, microenvironment concentrations and the results of ambient fixed site monitoring.

Extensive work resources and monitoring equipment are, however, required to carry out the measurements to obtain, for example, the residential outdoor concentrations of pollutants or the total personal exposures of the urban population. With the assistance of models, the results from exposure measurements and ambient fixed site measurements can be generalized to estimate the exposure of larger populations and used, for example, to evaluate scenarios in traffic planning.

The PM exposure problem is far more complicated than how well the ambient fixed site concentrations represent total personal exposures. Numerous outdoor and indoor sources produce particles and these particles exhibit different elemental and chemical compositions and size distributions. These characteristics define the penetration of particles into buildings, into lungs, and further, from lung alveoli to blood circulation. The particles are also assumed to be of different toxicity. The characteristics of particles which cause detrimental health effects remain a puzzle for researchers.

The aims of this thesis were:

- to determine spatial and temporal variations in $PM_{2.5}$ and NO_2 concentrations in the Helsinki Metropolitan Area
- to define whether $PM_{2.5}$ and NO_2 concentrations measured at ambient fixed sites can be used to evaluate total personal exposures
- to study how well regression models can evaluate total personal exposures
- to develop a model which combines the predicted concentrations, information on people's activity and the location of the population to evaluate personal exposures to ambient origin air pollution and further illustrate these results using the Geographic Information System (GIS)

2 Review of literature

2.1 Exposure assessment

Air pollution levels could have substantial temporal and spatial variation between different locations (e.g. indoor levels may be dissimilar to outdoor levels). People visit many different locations over the course of a single day (home, workplace, shops, etc.). Locations with a more or

less homogeneous pollutant concentration in time and space can be defined as microenvironments (Ott, 1980).

According to the United States Environmental Protections Agency (EPA, 1992), exposure to a pollutant is the contact of a chemical, physical or biological agent with the outer boundary of an organism. Exposure of an individual to a pollutant can also be defined as the contact concentrations of the pollutant experienced by the individual (Georgopoulos and Lioy, 1994) or as the coexistence of an individual and a pollutant in the same microenvironment (Ott, 1995).

Mathematically, exposure (E_i) for individual *i* can be defined as (Lioy, 1990):

$$E_i = \int_{t0}^{t1} c(t)\ \mathrm{d(t)} \qquad (1)$$

,where *c(t)* is a concentration of a pollutant, which varies as a function of time, and *t* is time, t_1-t_0 being the exposure duration. The integrated area under the curve of this profile is defined as the magnitude of the exposure (e.g. $\mu g/m^3 * h$).

The route of a contaminant from its source into the body is described Figure 1.

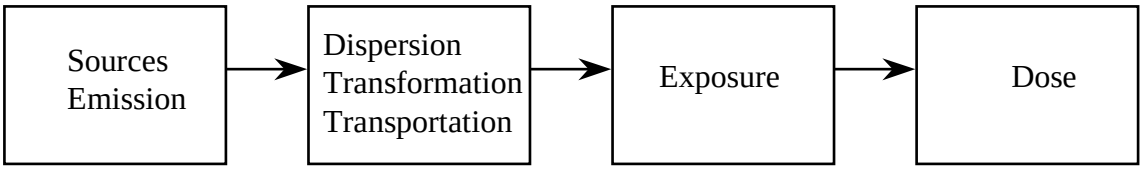

Figure 1. Exposure scheme.

The best method of assessing total personal exposure would be to obtain the full frequency distribution of minute values of an air pollutant, with the concentrations all being measured from the person as she/he moves through a series of microenvironments. This ideal is, however, very difficult to achieve in practice.

Total exposure can be assessed by using different direct and indirect indicators (Monn, 2001). In the direct approach, exposure levels are determined in individuals, and in the indirect approach either stationarily or by models.

Direct measurements

a) Personal exposure monitoring

Participants carry a device that samples the pollutant or registers pollutant levels during that person's normal daily activity as she/he moves from one microenvironment to another.

b) Biological monitoring

This involves using some type of bodily material (e.g. urine, saliva, blood) from which concentration of a pollutant or the metabolite of a pollutant is determined.

Indirect measurements

c) Ambient fixed site monitoring

Ambient fixed site data are usually obtained from routine ambient air quality monitoring stations. The exposure of the population is determined according to pollutant levels as measured at the nearest monitoring site or according to variation over time. It is often assumed that the exposure around a monitoring site is uniformly distributed. Concentrations measured at ambient fixed sites can only estimate exposures originating from outdoor sources.

d) Multi-microenvironment concentration measurements

Pollutant levels are measured in a few selected microenvironments (e.g. home and workplace) that are considered to have a major contribution to the integrated exposure based activity patterns and expected pollutant levels.

e) Models

Modelling can be based on, for instance, the qualitative or categorical classification of a population. The study population is classified according to a characteristic that distinguishes between low and high exposure, based on the proximity of pollutant sources. This characteristic can be the area of residence (rural vs. urban), job classification (occupational exposure) or the presence of an indoor pollutant source (e.g. gas cooking or smoking). All members in an exposure class are assumed to have the same or at least comparable exposures that are different from the exposure of members of a different exposure class. But the models can also be more complicated (see 2.5).

2.2 Ambient fixed site concentrations

$PM_{2.5}$

Airborne particulate matter represents a complex mixture of organic and inorganic substances. The size of atmospheric aerosol particles are considered to range from a few nanometres to tens of

micrometres in diameter. Particles less than 2.5 μm in diameter are generally referred to as “fine“ and those greater than 2.5 μm diameter as “coarse“. However, as can be seen from Figure 2, the cut-off between fine and coarse particles should be 1 μm rather than 2.5 μm. This way, the fine and coarse modes could be more clearly characterized through the different processes acting on them, i.e. the mechanisms by which they are formed and transformed in, and removed from, the atmosphere. They also require different techniques for their removal from sources, have different chemical compositions and optical properties, and differ significantly in their deposition patterns in the respiratory tract.

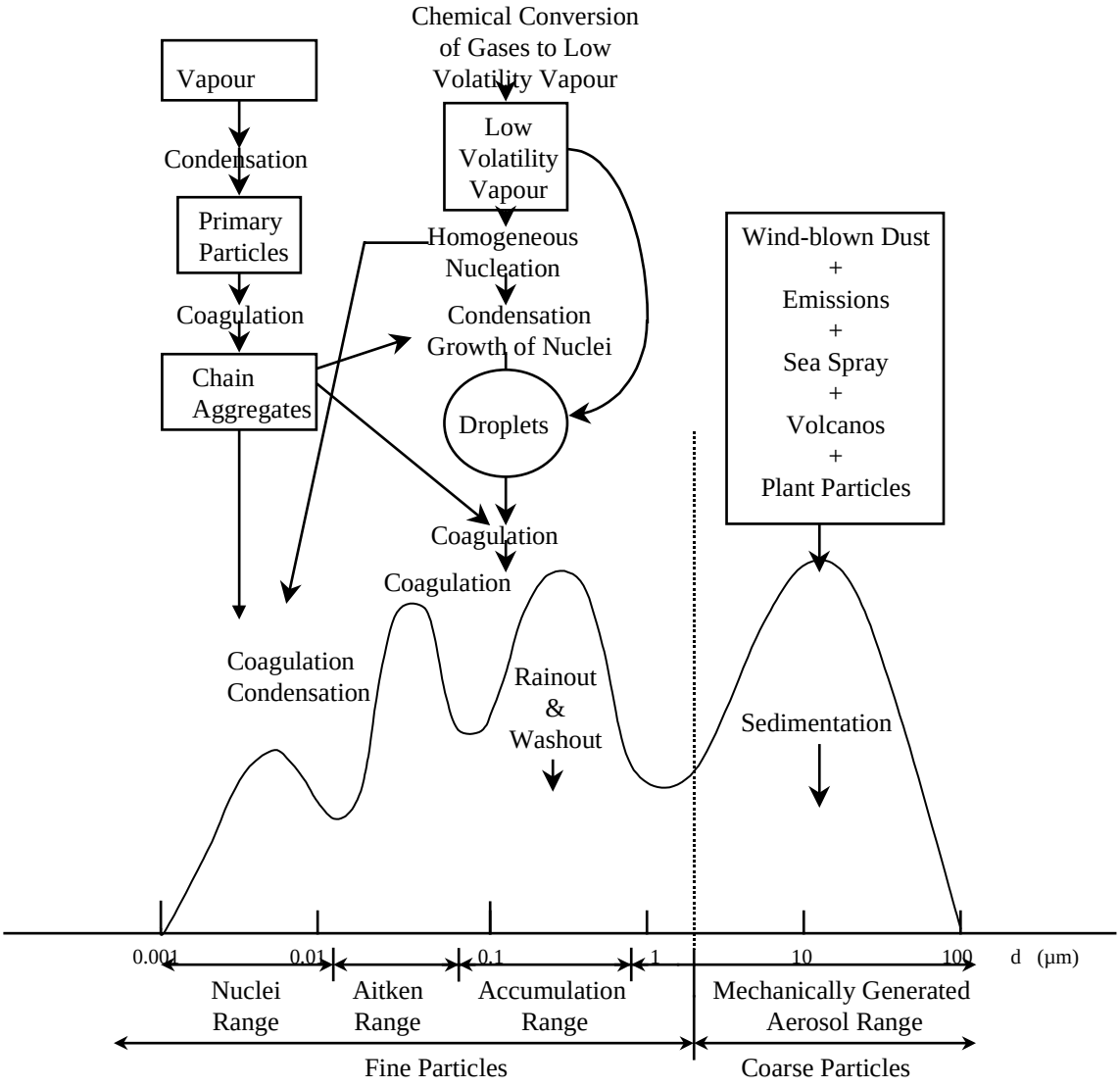

Figure 2. Idealized schematic of the distribution of the particle surface area of an atmospheric aerosol. Principal modes, sources, and particle formation and removal mechanisms are indicated (modified from Seinfield and Pandis, 1998).

Fine particles can roughly be divided into three modes: the nucleation mode, the Aitken mode and the accumulation mode. The nucleation mode (less than approximately 20 nm) and the Aitken mode

(approximately 20 - 90 nm) together account for the majority of particles. However, because of their small size, these particles rarely amount to more than a few percent of the total mass of airborne particles. Particles in the nucleation and the Aitken modes are formed from vapour condensation during combustion processes and from the nucleation of atmospheric species to form fresh particles. They are lost principally by coagulation with larger particles.

The accumulation mode containing particles from approximately 0.09 μm to 2.5 μm in diameter usually accounts for most of the aerosol surface area and a substantial part of the aerosol mass (or volume). The sources of particles in the accumulation mode are coagulation, condensational growth of particles in the Aitken mode and cloud processes. Because removal mechanisms that are efficient at the small and large particle extremes of the size spectrum are inefficient in the accumulation range, these particles tend to have considerably longer atmosphere residence times than those in the Aitken or coarse mode. The coarse mode, from > 2.5 μm in diameter, is formed by mechanical processes and usually consist of man-made and natural dust particles (Finlayson-Pitts and Pitts, 1986; Seinfield and Pandis, 1998).

Natural background concentrations of $PM_{2.5}$ are almost as high as they are in urban areas. For example, $PM_{2.5}$ concentrations have varied between 5.4 and 6.5 $\mu g/m^3$ in the rural background site of Hyytiälä, Finland, during 1999 and 2001 (Kulmala et al., 2001; Aalto, 2002). Pakkanen et al. (2001) measured $PM_{2.5}$ concentrations in urban traffic (Vallila), in semi-urban (Herttoniemi) and urban background (Luukki) sites in 1996-97. The average $PM_{2.5}$ concentrations were 11.8, 9.1 and 8.4 $\mu g/m^3$, respectively. The similarities in mass concentrations suggested that on average almost two-thirds of the $PM_{2.5}$ mass at the urban site is not of local origin. The trend also being similar at these three sites indicates that long-range transport dominates the $PM_{2.5}$ mass concentrations.

NO_2

Nitrogen dioxide (NO_2) is one of the major gaseous air pollutants in urban air. It is soluble in water, reddish-brown in colour and a strong oxidant. NO_2 is an important atmospheric trace gas because 1) it absorbs visible solar radiation and contributes to impaired atmospheric visibility, 2) as an absorber of visible radiation, it could have a potential direct role in global climate change if its concentrations were to become sufficiently high, 3) it is, along with nitric oxide, the chief regulator of the oxidizing capacity of the free troposphere by controlling the build-up and fate of radical species, including hydroxyl radicals, and 4) it plays a critical role in determining ozone concentrations in the troposphere because the photolysis of nitrogen dioxide is the only key initiator

of the photochemical formation of ozone, whether in polluted or unpolluted atmospheres (WHO, 2000).

On a global scale, emissions of nitrogen oxides from natural sources far outweigh those generated by human activities. Natural sources include intrusion of stratospheric nitrogen oxides, bacterial and volcanic actions, and lightning. Because natural emissions are distributed over the entire surface of the earth, however, the resulting background atmospheric concentrations are very small. For example, the annual average NO_2 concentrations have varied between 6 and 10 $\mu g/m^3$ in the urban background site of Luukki during 1993 and 2000 (Aarnio et al., 2001). The annual average NO_x (NO_2 + NO) concentrations have varied between 1.8 and 2.3 ppb (about 3-4 $\mu g/m^3$) in the rural background site of Hyytiälä in1997 and 2001 (Kulmala et al., 2001; Aalto, 2002). The major source of anthropogenic emissions of nitrogen oxides into the atmosphere is the combustion of fossil fuels in stationary sources (heating, power generation) and in motor vehicles (internal combustion engines). Urban outdoor levels vary according to the time of day, season and meteorological factors. Indoor sources, such as cooking with gas or cigarette smoking, may be the main contributors to individual exposure (WHO, 2000).

2.3 Microenvironment concentrations and major indoor sources

People in developed countries spend most of their time (from 80% to 90%) indoors (e.g. Schwab et al., 1990; Jenkins et al., 1992; Ackermann-Liebrich et al., 1995). Residential and workplace indoor levels therefore have a very large effect on total personal exposures.

$PM_{2.5}$

In most studies, the participants of the measurements have been volunteers. Only a few population probability-based PM studies exist. The PTEAM (Particle Total Exposure Assessment Methodology) study was the first large-scale population probability-based PM exposure study (Clayton et al., 1993; Thomas et al., 1993; Özkaynak et al., 1996). Total personal PM_{10} exposures of non-smokers and PM_{10} and $PM_{2.5}$ levels were measured indoors and outdoors in Riverside, California, in the autumn of 1990. Another large probability-based $PM_{2.5}$ exposure study was the RTI (Research Triangle Institute) study, where the three-day average total personal $PM_{2.5}$ exposures and residential indoor and outdoor levels in Toronto, Canada, from June 1995 to August 1996 were measured (Pellizzari et al. 1999).

Cigarette smoking is an important indoor source of $PM_{2.5}$. Smoking increased indoor concentrations by 27 μg/m^3 during the day and 32 μg/m^3 during the night in the PTEAM study (Özkaynak et al., 1996). In the EXPOLIS study, mean total personal exposure concentrations of active smokers (31.0 ± 31.4 μg/m^3) were almost double those of participants exposed to environmental tobacco smoke (16.6 ± 11.8 μg/m^3) and three times those of participants not exposed to tobacco smoke (9.9 ± 6.2 μg/m^3) in Helsinki, Finland (Koistinen et al., 2001).

Abt et al. (2000a,b) have studied indoor sources and particle size distribution (from 0.02 to 10 μm) in non-smoking households. According to their model, cooking, cleaning and indoor work had a strong influence on indoor particle concentrations, primarily on particles larger than 2 μm. Cleaning appeared to have little effect on indoor particles, with the exception of coarse particles ranging from 5 to 10 μm. Cooking performed in the previous 20-min period also significantly increased concentrations for 0.02-0.5 μm particles. Similar results were obtained by Raunemaa et al. (1989) in their measurements in Helsinki. Cooking and frying increased concentrations of both fine and coarse particles, but because this kind of particle production was found to be infrequent, its contribution to the average indoor concentration was minor. Cooking added 13 μg/m^3 (25% of total indoor concentration) to daytime indoor $PM_{2.5}$ concentrations but was not significant during overnight periods in the PTEAM study (Özkaynak et al., 1996). Jones et al. (2000), by contrast, reported that cooking has only a slight effect on indoor $PM_{2.5}$ concentrations. Cleaning had no influence on indoor particle concentrations in the PTEAM study. Abt et al. (2000b), however, found that cleaning was one of the indoor sources of $PM_{2.5}$ concentrations. Differences in the time resolution of sampling methodologies used in their study (20 min) and in the PTEAM study (12 h) may contribute to differences in source contributions.

Room-to-room variation of $PM_{2.5}$ has generally been found to be low (difference between livingroom and kitchen less than 10% when mass integrated over a 12 h period), even in the presence of smoking activity or gas cooking (e.g. Wigzell et al., 2000).

Ambient fixed site concentrations have explained quite well the variation in residential outdoor concentrations (r^2=0.67-0.92) (Clayton et al., 1993; Pellizzari et al., 1999). Leaderer et al. (1999) measured homes in Virginia, USA, and although homes were located as far as 175 km from the regional ambient fixed site, an association (r^2=0.24) was present between residential outdoor and ambient fixed site concentrations during summer months.

Associations between for the residential indoor and outdoor concentrations have been observed to vary markedly (r^2=0.05-0.76) because of considerable variation in the amount of indoor sources (Clayton et al., 1993; Garret et al., 1999; Leaderer et el., 1999; Pellizzari et al., 1999; Abt et al., 2000a,b; Jones et al., 2000; Lachenmyer and Hidy, 2000; Patterson and Eatough, 2000).

Ratios between indoor and outdoor concentrations (I/O) have been higher in the presence of smoking and gas appliance use indoors, varying between 0.54 and 1.08 for residents without these major PM sources (Li, 1994; Monn et al., 1997; Jones et al., 2000; Morawska et al., 2001) and between 1.0 and 1.8 for those in smoking and gas appliance environments (Monn et al., 1997; Brauer et al., 2000).

Associations between residential indoor and ambient fixed site concentrations were quite high in the PTEAM study (day: r^2=0.42 and night: r^2=0.55) but considerably lower in the RTI study (r^2=0.04) (Clayton et al., 1993; Pellizzari et al., 1999).

NO_2

The most important indoor sources of NO_2 are different kinds of gas appliances such as gas stoves, gas ovens, gas space and water heaters, and unvented kerosene heaters. NO_2 concentrations in homes with gas appliances are usually 1.5-5 times higher than in homes with electric stoves (Spengler et al., 1994; Alm et al., 1998; Levy et al., 1998; Monn et al., 1998; Garret et al., 1999; Chao and Law, 2000; Lee et al., 2000).

Some studies have shown that also tobacco smoking adds only marginally (1-4 $\mu g/m^3$) to NO_2 concentrations (Alm et al., 1998; Levy et al., 1998; Monn et al., 1998; Garret et al., 1999). However, according to Lee et al. (2000), tobacco smoking can have an even larger effect than the previously mentioned factors on NO_2 concentrations; the presence of a smoker increased mean indoor NO_2 levels ca. 1.5-fold the mean indoor levels without smokers.

A couple of large NO_2 exposure studies, namely the Swiss SAPALDIA (Swiss Study on Air Pollution and Lung Diseases in Adults, Monn, et al., 1998) and the Los Angeles Basin (Spengler et al., 1994) studies, have been conducted. Residential indoor and outdoor levels and total personal exposures to NO_2 were determined for more than 500 participants in a subpopulation of SAPALDIA. In the Los Angeles Basin study a representative sample of 482 households and 682 individuals was measured in the one-year period between 1987 and 1988.

The associations between indoor and outdoor concentrations were at the same level (r^2= 0.34, SAPALDIA; r^2=0.41, Los Angeles Basin) for residences without gas cooking. In the Los Angeles Basin study associations were a little bit lower (r^2=0.39) with gas cooking but higher with gas with pilot lights (r^2=0.44), however, this difference was not significant. Spengler et al. (1994) suggested that this difference might actually reflect the variation in ambient NO_2 concentrations across the Los Angeles Basin.

The I/O ratios have varied from 0.40 to 0.80 for residences without ETS (environmental tobacco smoke) or gas appliances, and from 0.80 to 3.2 for residences with gas stoves and/or smoking (Quackenboss et al., 1986; Monn et al., 1997; Levy et al., 1998; Garret et al., 1999; Heal et al., 1999; Chao and Law 2000; Lee et al., 2000).

2.4 Personal exposure

Personal exposures can be measured cross-sectionally (between individuals) or longitudinally (within individuals). This thesis is based on the EXPOLIS study, which is a cross-sectional study. Each participant was measured only once, only one to four participants were measured at the same time and the measurement period was about 48-h.

$PM_{2.5}$

Total personal exposures are usually higher than ambient fixed site or microenvironment concentrations (e.g. Brauer et al., 2000; Rojas-Bracho et al., 2000) because people are exposed to microenvironments that are not measured such as commuting and restaurants. Some studies have reported that a $PM_{2.5}$ personal cloud might exist (according to Williams et al. (2000), 3.1 $\mu g/m^3$ for elderly participants). Other studies have reported only a weak or non-existent personal cloud (Lachenmyer and Hidy, 2000)

In the RTI study, the association between total personal exposure and ambient fixed site concentration was lowest (r^2=0.04), that between total personal exposure and residential outdoor concentration was slightly higher (r^2=0.05), and that between total personal exposure and residential indoor concentration was highest (r^2=0.62) (Pellizzari et al., 1999). Lachenmyer and Hidy (2000) also found higher associations between total personal exposures and residential indoor concentrations (r^2=0.22 and 0.64 in summer and winter, respectively) than between total personal exposures and residential outdoor concentrations (r^2=0.12 and 0.51 in summer and winter, respectively). Janssen et al. (1999), however, reported a higher association between children's total

personal exposure and ambient fixed site concentration (r^2=0.17) than was observed in the RTI study.

NO_2

The typical outcome of NO_2 exposure studies is that NO_2 exposures are lower than ambient levels (Spengler at al., 1994; Alm et al., 1998; Chao and Law, 2000; Mukala et al., 2000). Indoor sources dominate total personal exposures. Therefore, total personal exposures were quite well explained by indoor concentrations (r^2=0.36-0.76) but less well explained by outdoor concentrations (r^2=0.04-0.51) (Quackenboss et al., 1986; Spengler et al., 1994; Monn et al., 1997; Levy et al., 1998; Heal et al., 1999; Rojas-Bracho et al., 2001).

2.5 Models of human exposure

Models for evaluating exposure to air pollutants have been classified as statistical, mathematical and mathematical-stochastic (modified from Ryan, 1991). The statistical approach involves the statistical determination of the measured exposures in terms of the factors that are assumed to influence these exposures (e.g. regression models). Mathematical modelling involves the application of emission inventories combined with atmospheric dispersion and population activity modelling (e.g. Clench-Aas et al., 1999; Jensen, 1999; Johansson, 1999). The stochastic approach attempts to include a treatment of inherent uncertainties in the model, such as those caused by the turbulent nature of atmospheric flow (for example, Monte-Carlo simulations, e.g. Hänninen et al., 2002).

Ambient air pollution concentrations can be computed by dispersion models. To evaluate the spatial distributions of exposures, air pollution concentrations can be combined with the population statistics (e.g. Brauer et al., 2001). Some studies have utilized additional time-microenvironment data (Clench-Aas, 1999; Jensen, 1999) or divided the entire population into night and day populations according to statistics on homes and workplaces (Johansson et al., 1999).

In most studies, the indoor concentrations are evaluated by multiplying the outdoor concentrations by a constant (as in Paper V; Jensen, 1999; de Haan, 2001). Dimitroupoulou et al. (2001), however, have estimated the indoor concentrations by an indoor model evaluating NO_2 exposure. Kulmala et al. (1999) have developed an indoor model for particles. A drawback for models of this kind, though, is the difficulty in obtaining exact building-specific input parameters (e.g. ventilation rate, indoor volume, indoor total surface area, penetration coefficient).

3 Materials and methods

3.1 Study design

3.1.1 EXPOLIS

EXPOLIS (air pollution exposure distributions within adult urban populations in Europe) was a multi-centre exposure study in Europe (Centres: Athens, Greece; Basel, Switzerland; Grenoble, France; Helsinki, Finland; Milan, Italy; Prague, Czech Republic; and later also Oxford, UK) (Jantunen et al., 1998, 1999). The purpose of the EXPOLIS study was to measure the exposures of adult urban populations in Europe to major air pollutants and some of the key parameters affecting these exposures. A further aim was to improve environmental health risk management by developing a technique for assessing and predicting air pollution exposure consequences of alternative urban development policies. The EXPOLIS study focused on adult (25-55 years) European urban populations and their personal air pollution exposures to nitrogen dioxide (NO_2) (Paper III; Rotko et al., 2001), carbon monoxide (CO), fine particles ($PM_{2.5}$) (Paper II; Rotko et al., 2000a; Koistinen et al, 2001, 2002) and 30 volatile organic compounds (VOCs) (Edwards et al., 2001a,b,c; Jurvelin et al., 2001a,b).

CO and NO_2 represent exposure to traffic exhaust and indoor combustion sources. The VOCs were selected as study compounds because of health and welfare concerns both indoors and outdoors: many VOCs are useful as source markers, and present data on VOCs are of variable quality. In addition, while $PM_{2.5}$ is of great health concern and interest today, very few $PM_{2.5}$ exposure studies on representative population samples have been reported thus far. The working-aged urban populations were selected because their exposures are most affected by urban traffic planning, zoning and occupational conditions, i.e. they would provide the most variable data for exposure model development and validation.

Time-activity data were also collected by a time-activity diary with a 15-min resolution. Information on other possible factors affecting personal exposures was collected using a questionnaire, which was completed by each participant at the end of the measurement period.

The study was conducted from summer 1996 to winter 1997-98 during weekdays. In Helsinki, 201 participants were measured, and time-activity data and questionnaires were collected from 234 participants. The population sample was representative of the target population in Helsinki (Rotko et al., 2000b). Fifty participants were measured in the other EXPOLIS centres, and time-activity and questionnaire data were collected from 50 (Athens and Prague) to 250 (Basel) participants.

Although the response rate in Basel was only 49%, the sample population was fairly representative of the target population of Basel.

In Athens, the participants were volunteers and they were more educated and smoked less than the overall population in Athens. In Prague, the participants (also volunteers) were selected from a restricted area (Prague V in the city centre), and they were considerably younger, better-educated and smoked less than the target population. The population sampling and sample quality issues are described in detail in Rotko et al. (2000b) and Oglesby et al. (2000).

3.1.2 EXPAND

The EXPAND (EXPosure to Air pollution, especially to Nitrogen Dioxide and particulate matter) project aims at the development of a population exposure model for ambient nitrogen dioxide (NO_2) and the fine particles ($PM_{2.5}$). This includes the refinement of the existing atmospheric dispersion model for nitrogen oxides (Karppinen et al., 2000) and the development of a dispersion model for $PM_{2.5}$. The computed microenvironment concentrations and exposures were compared with data from the EXPOLIS study. The predicted concentrations were also compared with the Helsinki Metropolitan Area Council (YTV) air quality monitoring results.

3.2 Study locations

This thesis presents $PM_{2.5}$ and NO_2 results from the following EXPOLIS centres: Athens, Basel, Helsinki Metropolitan Area and Prague. Table I presents the population number, average temperatures in winter and summer and the annual average rainfall in these centres.

Table I. Population, average temperatures in January and July and the annual average rainfall in Greater Athens, Basel, Helsinki Metropolitan Area and Prague.

City	Population number	Temperature (°C)		Rain (mm)
		January	July	
Athens	3 000 000	+10	+28	400
Basel	400 000	+1	+18	1 000
Helsinki	900 000	-6	+17	700
Prague	1 200 000	-1	+19	500

Athens and Prague are the capitals and the largest cities of Greece and the Czech Republic, respectively. The Helsinki Metropolitan Area is the largest urban agglomeration in Finland

comprising four cities: Helsinki (the capital), Espoo, Vantaa and Kauniainen. Basel is a major centre of chemical and pharmaceutical industries and a commercial port in Switzerland.

3.3 Monitoring methods

3.3.1 Personal and microenvironment measurements

$PM_{2.5}$

The $PM_{2.5}$ exposure was measured with a personal exposure monitor (PEM). Each participant carried the sampler for a sampling period of 48-h. The PEM consisted of an air pump (Buck IH pump, A.P. Buck Inc. Orlando, FL, USA), a cyclone (GK2.05, BGI Inc. Waltham, MA, USA) and a filter holder (37-mm Millipore filter holders, Millipore Corporation, Bedford, MA, USA) with a Gelman Teflo filter (2-µm pore size). Two filter holders were provided for each participant: one 'workday filter' to be used for the time periods beginning when leaving home for work and ending when returning home from work (ca. 2 x 8-10 h), and one 'leisure-time filter' for the remaining times (ca. 2 x 14-16 h).

Microenvironmental monitors (MEMs) were placed at participants' homes outdoors and indoors and in their workplaces (indoors) for 48-h to collect microenvironment $PM_{2.5}$. The pumps were programmed to run at home both outdoors and indoors during the expected non-working hours and at the workplace during the expected working hours of each participant. The MEM sampler contained an EPA-WINS impactor (EPA Well Impactor Ninety-Six, BGI Inc. Waltham, MA, USA), a 47-mm filter holder (BGI) with a Gelman Teflo filter and a PQ100 pump (BGI). Quality assurance and quality control (QA/QC) procedures and methods of $PM_{2.5}$ sampling are presented in detail in Koistinen et al. (1999).

NO_2

NO_2 concentrations were measured by passive Palmes tubes (Palmes, 1976). NO_2 tubes were fixed on PEM and MEM monitors. In addition to collecting personal exposures, the tubes were open throughout the measurement period (about 48-h) indoors (both at home and workplace) as well as outdoors. NO_2 was measured as nitrite in all samples from all centres using a spectrophotometric method at the Institute for Hygiene and Applied Physiology, ETHZ, Zürich, Switzerland (Monn et al., 1998). NO_2 concentrations were calculated according to Fick's law (Bird et al., 1960).

3.3.2 Ambient fixed site monitoring

In the Helsinki Metropolitan Area, the network contains nowadays six permanent multicomponent stations. These are located at the Helsinki city districts of Töölö, Vallila and Kallio, in the district centres of Leppävaara and Tikkurila, in Espoo and Vantaa, respectively, and in Luukki, the rural area of Espoo. Mobile stations are re-located each calendar year. The $PM_{2.5}$ measurements commenced at the Vallila site in March 1997, and at the Kallio 2 site in the beginning of 1999 (Aarnio et al., 2001).

3.4 Quality assurance

Performance criteria for the quality assurance (QA) program in EXPOLIS were generally to minimize any differences between the centres which would affect comparability of results and to ensure quantified data for all exposures and microenvironment concentrations. Quality assurance was based on the principle that all procedures must be carefully planned, tested, documented and performed according to standard operating procedures (SOPs).

For NO_2 QA, laboratory blanks (randomly selected tubes kept closed in the laboratory during shipment, storage and sampling with other tubes in the same batch), shipment blanks (tubes shipped with sampling tubes but kept closed the entire time) and field blanks (tubes followed with the sampling tubes also during the measurement but kept closed the entire time) were used. Similar blanks were also used for $PM_{2.5}$ filters. Duplicate samples were used for each step, from preparation to sampling and analysis.

The average difference between the field NO_2 duplicates was 2.9 $\mu g/m^3$ (n=167, Helsinki) 4.2 $\mu g/m^3$ (n=77, Basel) and 10.6 $\mu g/m^3$ (n=21, Prague, 2.8 $\mu g/m^3$ if three outliers are excluded). The average of shipments blanks was 0.10 $\mu g/m^3$ and the average absorbance of laboratory blanks varied between 0.00 and 0.01 $\mu g/m^3$ for batches in all centres (Paper III, Rotko et al., 2001).

The average absolute difference for $PM_{2.5}$ PEM duplicates was 2.1 $\mu g/m^3$ and the standard deviation was 2 $\mu g/m^3$, the corresponding figures for MEM duplicates were 0.7 $\mu g/m^3$ and 0.6 $\mu g/m^3$, respectively. The field blank filters showed systematic mass increase during the field measurements. The average mass increase and standard deviation were 5.8 μg and 5.1 μg for MEMs (n=74), and 1.8 μg and 4.6 μg for PEMs (n=66) during one week (Koistinen et al., 1999).

The accuracy of continuous PM monitoring devices depends on the PM composition and local meteorological conditions, due to possible evaporation of semi-volatile material. The daily average

concentrations of PM_{10} and $PM_{2.5}$, determined by the Eberline FH 62 I-R analysers, were therefore compared with corresponding results obtained by virtual impactors. These comparisons were performed at the station of Vallila, Helsinki, from June 1999 to May 2000. The comparisons indicated a good agreement of results. For the concentrations of $PM_{2.5}$, the slope of the regression line (k) was 0.98, the constant (c) was +0.03 μg m^{-3} and r^2 was 0.91 (Aarnio et al., 2001). Field intercomparisons were also conducted, in which the concentrations of PM_{10} obtained by TEOM monitors were compared with those obtained by other methods; these also indicated a good agreement of results (Sillanpää et al., 2001).

3.5 Data analyses

3.5.1 Statistical analyses

Data were analysed using the STATA statistical package version 5.0 (Stata Corporation, College Station, Texas, USA) as presented in Papers II and III. Because the $PM_{2.5}$ and NO_2 data distributions were positively skewed, log-transformed data were used in these analyses.

To compare the predicted and measured NO_2 concentrations, the index of agreement and fractional bias were used (Paper IV), and are defined as follows (Willmot, 1981; Petersen, 1997):

$$\text{the index of agreement, } IA = 1 - \frac{\overline{(C_P - C_O)^2}}{\left[\overline{\left|C_P - \overline{C_O}\right| + \left|C_O - \overline{C_O}\right|}\right]^2}, \qquad (2)$$

$$\text{the fractional bias, } FB = \frac{\overline{C_P} - \overline{C_O}}{0.5\left(\overline{C_P} + \overline{C_O}\right)}, \qquad (3)$$

,where C_P and C_O are the predicted and observed concentrations, respectively. The overbar refers to the average over of hourly values. *IA* is a measure of the correlation of the predicted and observed time series of concentrations, while *FB* is a measure of the agreement of the mean concentrations.

3.5.2 Modelling

Time-weighted microenvironmental exposure model (TWME)

Time-weighted average NO_2 exposures were estimated based on residential indoor and outdoor, workplace and ambient fixed site concentrations and time-microenvironment activity patterns according to the following equation (Paper III):

$$P_i = \Sigma t_{ij} M_{ij} / t_{total} \quad (4)$$

,where j is the microenvironment (residential indoor, residential outdoor, workplace indoor and ambient fixed site concentrations), i is the participant index, P_i is the estimated time-weighted average personal NO_2 exposure for participant i, M_{ij} is the NO_2 concentration for participant i in the microenvironment j, t_{ij} is the number of hours spent in microenvironment j for participant i during the sampling period.

Regression models

Personal NO_2 exposures were modelled using a log-linear multiple regression model according to the following equation (Paper III):

$$\ln(P_i) = \Sigma \alpha j C_{ij} + \ln(O_i) \quad (5)$$

,where P_i is the measured personal NO_2 exposure for participant i, C_{ij} is the indicator variable for j^{th} characteristic for individual I, α_{ij} is the calculated regression coefficient for j^{th} individual characteristic, O_i is the measured residential outdoor or ambient fixed site concentration for participant i.

EXPAND

The exposure assessment of ambient origin air pollution (more details in Paper V; Kousa et al., 2001) was divided into five phases: 1) emission inventory, 2) dispersion modelling, 3) location and time activity of population modelling, 4) combination of predicted concentrations and population data and 5) presentation of results by geographic information system (GIS).

1) Traffic flows and travel speeds were computed using the EMME/2 transportation system (INRO, 1994). Emissions were evaluated using the EMME/2 system and emission factors that have been evaluated for this area (Helsinki Metropolitan Area Council, 1997). The model allows for diurnal and weekly variations both in traffic volumes and speeds, as well as in traffic emissions. Emission inventory of NO_x had been updated in the Helsinki Metropolitan Area in 1996 and 1997 (Karppinen et al., 2000). The inventory included the emissions from various mobile sources (harbours and marine traffic, and aviation) and stationary sources (power plants, other point sources and residential heating). Stationary sources were considered as point or area sources.

2) The dispersion model is based on a combined application of the Urban Dispersion Modelling system (UDM-FMI, Karppinen et al., 1998) and the road network dispersion model (CAR-FMI, Härkönen et al., 1995, 1996). The relevant meteorological parameters for the models were evaluated using data produced by a meteorological pre-processing model (MPP-FMI) that has been specifically adapted for the urban environment (Karppinen et al., 1997, 1998, 2000). The modelling system includes a statistical and geographical analysis of the computed time series of concentrations. The modelling system also contains a method for the modelling of chemical interaction of pollutants originating from a large number of urban sources (Karppinen et al., 2000).

3) The population location and number in homes and workplaces were evaluated from an annually collected dataset (SeutuCD) in the Helsinki Metropolitan Area. Information for other locations, was obtained from a questionnaire collected by the Helsinki Metropolitan Area Council (1997). Other activity was evaluated using the questionnaire data together with information of journeys attracted by various places (e.g. shops) (Helsinki Metropolitan Area Council, 1990). The location of people in traffic was evaluated using the computed traffic flow information. This information was available separately for buses and cars for each street section on an hourly basis. Time-microenvironment activity data was combined with the location of the population in different microenvironments during the day as the activity of the population in different microenvironments.

4) Population data and the predicted outdoor concentrations (for simplicity, we assumed in our first version of the model that the residential and workplace indoor concentrations were the same as the corresponding outdoor concentrations) were combined using a model named EXPAND.

5) Finally, concentrations, activities and exposures to ambient air pollution were converted into a numerical format directly transferable to the GIS MapInfo system, and the results were presented as maps (we selected a grid size of 100 m * 100 m).

4 Measured and modelled concentrations and exposures

4.1 Ambient fixed sites in the Helsinki Metropolitan area

The $PM_{2.5}$ concentrations measured at the ambient fixed sites varied diurnally, being fairly uniform during working days, except for a moderate increase in the morning rush hour. Spatial variation was also moderate between the two measurements sites (Vallila and Kallio 2) (Paper I; Aarnio et al., 2001). The mean concentration varied between 8 $\mu g/m^3$ (Kallio 2 and Vallila in 2000) and 11 $\mu g/m^3$ (Vallila in 1998 and 1999), and the maximum day concentrations between 24 $\mu g/m^3$ (Kallio 2 in

2000) and 37 μg/m^3 (Vallila in 1999). In addition, in EXPOLIS, the ambient fixed site concentrations explained the variation of the residential outdoor concentrations quite well (r^2=0.81) in the Helsinki Metropolitan Area (Paper II). This is consistent with the fine PM variation reported for example, for the city and region of Basel (Röösli et al., 2000).

By contrast, the NO_2 concentrations varied both temporally and spatially: the mean concentrations varied between 17 μg/m^3 at Laaksolahti (1997) and 41 μg/m^3 at Töölö (1996), and the maximum day concentrations between 27 μg/m^3 at Luukki (2000) and 143 μg/m^3 at Sörnäinen (1996) (Paper IV; Aarnio et al., 2001). In addition, in EXPOLIS, the ambient fixed site concentrations explained the variation of the residential outdoor concentrations (r^2=0.20) in the Helsinki Metropolitan Area rather poorly (Paper III).

One reason for the difference between the spatial and temporal variation of $PM_{2.5}$ and NO_2 might be that their major outdoor sources are different. A large portion of $PM_{2.5}$ originates from long-range transport, while NO_2 is derived mostly from local traffic emissions in the Helsinki Metropolitan Area (Karppinen et al., 2000; Pakkanen et al., 2001; Koistinen et al., 2002).

4.2 Microenvironment concentrations and personal exposures

4.2.1 I/O ratio

Table II presents the I/O ratios for $PM_{2.5}$ for residents when there was no one smoking indoors or any gas appliances used, and for residents with smoking or gas appliances.

When the main indoor sources were excluded, the mean I/O ratio was highest (1.16) in Helsinki, where both indoor and outdoor concentrations were lowest, and lowest (0.83) in Athens, where the concentrations were highest. Buildings are designed, built and operated to isolate the occupants from the outdoors and accomplish this task to a varying extent. buildings are likely to be better isolated and ventilation lower in Helsinki (cold winter) than in Athens (mild winter). Therefore, the presence of indoor sources, such as smoking and gas appliances, increased the mean I/O ratio over twofold in Helsinki but only moderately in Athens.

Table II. Arithmetic means (average), number of measurements (n), geometric means (GM) and standard deviations (GSD) of the I/O ratios for $PM_{2.5}$ for all centres with and without major indoor sources.

$PM_{2.5}$	All	No smoking and no gas appliances indoors	Smoking and/or gas appliances indoors
		Helsinki	
average (n)	1.47 (162)	1.16 (143)	3.76 (19)
GM (GSD)	1.09 (1.91)	0.99 (1.66)	2.23 (2.99)
		Basel	
average (n)	1.69 (45)	1.07 (26)	2.55 (19)
GM (GSD)	1.17 (2.28)	0.96 (1.54)	1.52 (2.42)
		Prague	
average (n)	1.46 (20)	0.96 (7)	1.74 (13)
GM (GSD)	1.10 (2.05)	0.90 (1.51)	1.29 (2.13)
		Athens	
average (n)	1.05 (44)	0.83 (24)	1.31(20)
GM (GSD)	0.94 (1.60)	0.78 (1.48)	1.17 (1.59)
		Total	
average (n)	1.44 (271)	1.10 (200)	2.38 (71)
GM (GSD)	1.07 (1.89)	0.95 (1.62)	1.52 (2.35)

Table III presents the I/O ratios of NO_2 for residents with no one smoking indoors and no gas appliances, and for residents with the previously mentioned indoor sources.

Table III. Arithmetic means (average), number of measurements (n), geometric means (GM) and standard deviations (GSD) of the I/O ratios for NO_2 at all the centres with and without major indoor sources.

NO_2	All	No smoking and no gas appliances indoors	Smoking and/or gas appliances indoors
		Helsinki	
average (n)	0.76 (159)	0.73 (141)	0.95 (18)
GM (GSD)	0.67 (1.68)	0.65 (1.75)	0.82 (1.73)
		Basel	
average (n)	0.80(49)	0.69 (29)	0.96 (20)
GM (GSD)	0.71 (1.67)	0.62 (1.64)	0.87 (1.58)
		Prague	
average (n)	0.75 (29)	0.53 (5)	0.79 (24)
GM (GSD)	0.62 (1.95)	0.44 (1.95)	0.66 (1.94)
		Total	
average (n)	0.76 (237)	0.72 (175)	0.89 (62)
GM (GSD)	0.67 (1.68)	0.64 (1.72)	0.77 (1.77)

For NO_2, all mean I/O ratios were below 1. In addition, when the main indoor sources were excluded, the mean I/O ratio was highest (0.74) in Helsinki, where both indoor and outdoor concentrations were lowest, and lowest (0.53) in Prague, where the concentrations were highest (note, however, the small number of measurements in Prague). For NO_2 and $PM_{2.5}$, the presence of indoor sources had a greater effect on I/O ratios in Helsinki than in the other centres.

4.2.2 Associations

$PM_{2.5}$

Considering microenvironment concentrations of non-ETS-exposed participants in Helsinki, the highest association was obtained between the residential $PM_{2.5}$ outdoor and the ambient fixed site concentrations (r^2=0.85) and the weakest between the workplace indoor and ambient fixed site concentrations (r^2=0.15) (Paper II). In the PTEAM study, the association between nighttime ambient fixed and residential outdoor concentrations was higher (r^2=0.92), and in the RTI study, lower (r^2=0.67) than in Helsinki (Özkaynak et al., 1996; Pellizzari et al., 1999). Unfortunately, in the other centres, the ambient fixed site $PM_{2.5}$ concentrations were not measured continuously.

In Helsinki, the highest association with total personal $PM_{2.5}$ exposures was obtained with residential indoor concentrations during leisure time (r^2=0.80, ETS excluded), and the lowest with ambient fixed site concentrations during the workday (r^2=0.22, ETS excluded) (Paper II).

In the other centres, the highest correlation with total personal $PM_{2.5}$ exposures was obtained with either residential indoor concentrations during leisure time (with and without ETS exposure in Prague and without ETS exposure in Athens and Basel) or with workplace indoor concentrations during the workday (with ETS exposure in Athens and Basel). The weakest correlation was between total personal leisure-time exposures and residential outdoor concentrations (Paper II).

High correlations between total personal exposures and indoor concentrations are expected because the populations spend a great majority of their time inside their homes and workplaces, where the indoor concentrations are determined by PM penetrating from outdoor air, from indoor sources and from slow dilution by ventilation.

Exclusion of the ETS-exposed subpopulation had a two-way effect on the PM concentrations analysed. Removing the most significant indoor source unrelated to ambient air improved the correlations between personal (indoor) air and ambient (outdoor) air. On the other hand, inclusion of

the ETS-exposed group tended to increase the correlations between total personal exposures and indoor air concentrations as well as between total personal exposures during the workday and the leisure time. The reason for this was that including the ETS-exposed population in the analysis simultaneously broadened the ranges of both total personal exposures and indoor concentrations.

NO_2

In general, the respective correlations were lower for NO_2 than $PM_{2.5}$. The ambient fixed site concentrations explained only 16% of the residential outdoor NO_2 variation in Helsinki, 19% in Basel and 14% in Prague. The weakest association with NO_2 concentrations was found between the residential indoor and the ambient fixed site concentrations (centres combined: r^2=0.29). The association between the residential indoor and the residential outdoor concentrations was somewhat better (r^2=0.44, higher than in the SAPALDIA study, where r^2=0.37), and the association between total personal NO_2 exposures and residential indoor concentrations was highest (r^2=0.53, SAPALDIA: r^2=0.51) (Paper III; Monn et al., 1998).

The poorest association of total personal NO_2 exposures was found with ambient fixed site concentrations (r^2=0.19). Personal 48-h NO_2 exposures were associated more strongly with the residential outdoor and indoor concentrations (r^2=0.40 and 0.45, respectively). The strongest association was between 48-h personal exposures and workplace indoor concentrations (r^2=0.55) (Paper III). All NO_2 tubes remained open throughout the sampling period (about 48-h). This approach was the most problematic in the workplaces because participants spent only a fraction of the 48-h there. Moreover, many workplaces are located in areas where the outdoor activity level is quite different during off-work hours as compared with working hours, and in many workplaces, the ventilation is turned off during unoccupied hours. Consequently, workplace NO_2 measurements may not be very representative of concentrations during working hours.

Both $PM_{2.5}$ and NO_2 results were cross-sectional results in the EXPOLIS study. These results showed that short-term cross-sectional total personal exposures were poorly correlated with the respective ambient air concentrations. One reason for this is that multiple sources, in addition to ambient levels, exist participants' daily activities. The association between total personal exposures and ambient fixed site concentrations improved when environmental tobacco smoke (ETS) was not present and was better during leisure time than during working hours. However, longitudinal exposures correlate on average better than cross-sectional exposures with ambient fixed site concentrations. Janssen et al. (1999) in analysing children's $PM_{2.5}$ exposures with ambient fixed site concentrations, found the longitudinal association to be considerably better (r^2=0.74) than the

estimated cross-sectional association (r^2=0.17). Also the longitudinal association between total personal $PM_{2.5}$ exposures and ambient fixed site concentrations was high when elderly subjects with cardiovascular disease, both in Amsterdam and in Helsinki (r^2=0.62 and 0.58, respectively), was analysed (Janssen et al., 2000).

Fixed ambient site monitoring is most useful for following the day-to-day concentration variation and longer-term exposures. They may, however, be less valuable for assessing spatial variation, which is relevant for examining differences between total personal exposures of individuals. In general, the concentrations measured at ambient fixed sites better explain population rather than individual and long-term rather than short-term exposures.

NO_2 and $PM_{2.5}$

While associations between residential outdoor NO_2 and $PM_{2.5}$ were quite high in Basel (r^2=0.35) and somewhat lower in Helsinki (r^2=0.12), no association was found in Prague (r^2=0.00) (note the different sampling periods; NO_2 was measured for 48-h and $PM_{2.5}$ only during non-working hours). Associations between personal NO_2 and $PM_{2.5}$ exposures (both 48-h) were r^2=0.01, 0.11 and 0.14, when all measurements were included in Basel, Helsinki and Prague, respectively. When smoking indoors and use of gas appliances were excluded, this association increased in Helsinki and Prague (r^2=0.18 and 0.19, respectively) but not in Basel (r^2=0.00). As stated earlier, NO_2 and $PM_{2.5}$ concentrations have different outdoor sources and different spatial variation so it is not unexpected that the correlation between NO_2 and $PM_{2.5}$ exposures is weak.

4.3 Models

Personal NO_2 exposures were evaluated using three different models: the time-weighted microenvironmental exposure model (TWME), the regression model and the EXPAND model.

TWME model

Each total personal NO_2 exposure was estimated as the time-weighted average of that person's residential and workplace indoor, residential outdoor and ambient fixed site levels (using Equation 4). The model explained 70% of the measured total exposure variance (Paper III) in Helsinki and 74% when data from all centres were combined.

If all microenvironment concentrations and time-activity data were collected completely and accurately, the TWME model should duplicate total personal exposures. In practice, this is

presumably not achieved because only the most significant microenvironments are monitored. With appropriate microenvironment concentrations and time-activity data, however, the time-weighted microenvironmental model is fully generalizable and applicable to any population.

Regression models

Log-linear regression models using ambient fixed site concentrations combined with residential and workplace characteristics (work location, living in a high-rise building, building year, and keeping windows open (Equation 5)) explained 32% of the total personal NO_2 exposure variation. The corresponding regression model using residential outdoor concentrations explained 48% of this variation (Paper III) in Helsinki. When all centres were combined, a model based on residential outdoor concentrations explained the same 48% of the variation, whereas the ambient fixed site based model explained slightly more (37%) than for Helsinki alone.

The best regression model required microenvironment concentrations to be measured from inside and outside of residences and the workplaces of participants for whom the exposure was modelled (r^2=0.75). The same value was obtained for data from Helsinki alone and from all centres combined.

Every dataset produces a different regression model which can not generalized to a new setting (e.g. different populations, cities or time). This principle may be relaxed, however, if the regression model is based on sufficiently large and representative samples from different kinds of populations. Regression models are easy to use and do not need a lot of computation power.

EXPAND

Associations (r^2) between measured and predicted ambient fixed site hourly concentrations in Helsinki varied between 0.15 (Runeberg St., street canyon) and 0.58 (Tapiola, suburban residential) (Paper IV) in 1996 and 1997. The index of agreement (IA) (Equation 2) varied between 0.65 (Runeberg St.) and 0.82 (Töölö, urban traffic), while the fractional bias (FB) (Equation 3) values range from -0.29 (Laaksolahti, suburban residential) to +0.26 (Vallila, urban traffic).

Hourly time series of NO_2 concentrations of each participant in the EXPOLIS study were also modelled for the residential and workplace outdoor concentrations at the times when the participants were monitored. Modelled residential outdoor NO_2 concentrations explained 50% of the variation in measured residential outdoor NO_2 concentrations (Figure 3). The index of agreement was 0.74 and the fractional bias -0.29.

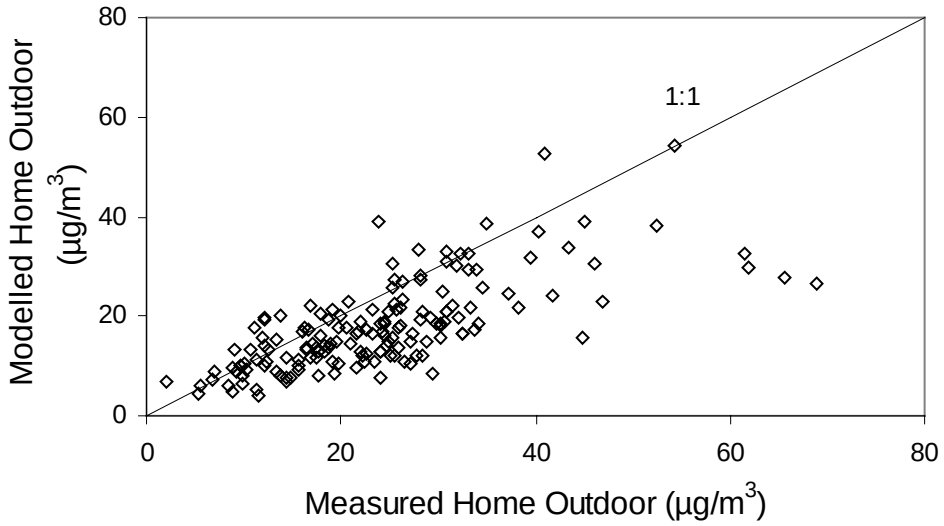

Figure 3. Modelled and measured residential outdoor NO_2 concentrations (r^2=0.50, n=161).

Generally, the modelled concentrations were lower than measured concentrations. The measured overall average residential outdoor concentration was 24 µg/m^3 (standard deviation, SD, was 11 µg/m^3), while the overall average modelled concentration was only 18 µg/m^3 (SD: 9 µg/m^3). On the other hand, according to Heal et al. (1999), short-term NO_2 measurements by passive samplers overestimated concentrations by 27%.

Figure 4. Modelled workplace outdoor NO_2 concentrations and measured workplace indoor NO_2 concentrations (r^2=0.35, n=121).

Modelled workplace outdoor concentrations explained 35% of the variation in measured workplace indoor NO_2 concentration (Figure 4). Naturally, this correlation was weaker than the above-mentioned residential correlation because the predicted concentrations were computed outside of the

workplaces and the measurements were done indoors (IA=0.52 and FB=-0.51), and the indoor sources were not included in the model.

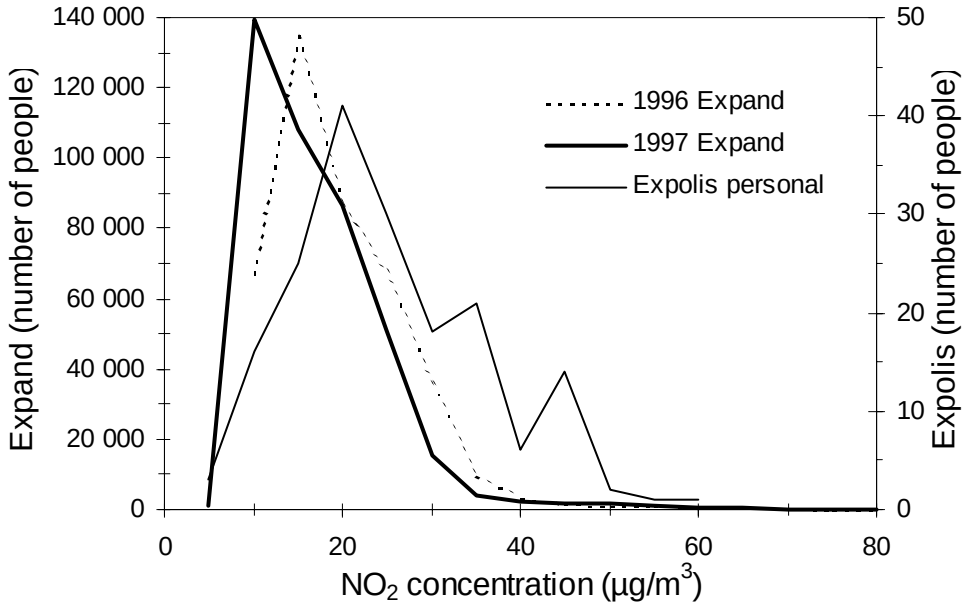

Figure 5. Predicted distributions of the number of people in combined microenvironments against the average concentration of NO_2 in 1996 and 1997 and the measured distribution of personal NO_2 exposure in the EXPOLIS study.

To compare the predicted exposures to ambient origin NO_2 and measured total personal exposures in EXPOLIS we computed the number of people against the average concentration of NO_2 at home, in the workplace, in other microenvironments and in traffic, and summed the number of people over these microenvironments. These results are shown in the same figure as the distribution of measured personal NO_2 exposure in the EXPOLIS study (Figure 5).

The modelled exposures to outdoor NO_2 are clearly lower than those measured. As mentioned earlier, the model underestimated the outdoor concentrations. Another reason for the discrepancy could be that indoor sources were not included in the modelled exposures.

Some sample results have been presented to illustrate the application of the model in Paper V. These include the concentrations, population densities, and exposures to ambient origin NO_2 in the Helsinki Metropolitan Area. Although people are exposed to the highest NO_2 concentrations in traffic, the time spent in traffic is relatively short compared with that spent at home and in work environments. Thus, the average exposure in traffic is clearly lower than that at home and in the workplace. The highest concentrations in traffic occur during the afternoon rush hours, and fairly high values also occur during the morning and evening.

The use of the EXPAND model provides spatial distributions of exposures to ambient origin air pollution and also enables evaluations of past and future scenarios. In principle, the model is applicable to any pollutant, so long as the required spatial concentration distributions are available. A model requires detailed input data, such as emission sources, meteorological data and time-activity data and running the dispersion model requires a lot of computation power.

5 Review of papers

This thesis describes associations between both $PM_{2.5}$ and NO_2 microenvironment concentrations and total personal exposures, and different ways to estimate the total personal exposure to air pollutants. Papers I and IV describe how $PM_{2.5}$ and NO_2 concentrations are dispersed across the Helsinki Metropolitan Area. Papers II and III describe the correlation between ambient fixed site concentrations, microenvironment concentrations and total personal exposures both for $PM_{2.5}$ and NO_2. Different kinds of regression models to estimate total personal NO_2 exposures are also examined in Paper III. Paper V describes an exposure model developed to estimate personal exposures to ambient origin air pollution. Paper IV also investigates how well the predicted NO_2 concentrations represent the measured NO_2 concentrations in the stations of the air quality measurement network.

Paper I examines $PM_{2.5}$ and PM_{10} concentrations measured in the air quality monitoring network in the Helsinki Metropolitan Area during 1997-1999. Temporal and spatial variation of the PM concentrations were studied. The evolution of urban PM concentrations in terms of relevant meteorological parameters during one PM episode were also studied. The concentrations of PM_{10} showed clear diurnal and spatial variations. In contrast, both spatial and temporal variations of $PM_{2.5}$ concentrations were moderate.

Paper II presents associations between total personal exposures, residential indoor, residential outdoor, workplace indoor and urban ambient air concentrations of $PM_{2.5}$ in four *EXPOLIS* centres (Athens, Greece; Basel, Switzerland; Helsinki, Finland and Prague, Czech Republic). Considering the whole chain from ambient fixed site to residential outdoor, residential indoor and total personal leisure-time (non-working hours) exposure, the correlations were highest between total personal exposures and residential indoor concentrations in all other centres except Athens. Ambient fixed site $PM_{2.5}$ concentrations were measured only in Helsinki. The residential indoor concentrations correlated quite well with ambient fixed site $PM_{2.5}$ concentrations, but data from ambient fixed site monitor was a poor predictor of total personal exposure to $PM_{2.5}$. It was a particularly poor predictor

of total personal workday exposures, and was considerably better for total personal leisure-time exposures. Log-linear regression models based on residential and workplace indoor concentrations of non-ETS-exposed participants explained over 2/3 of the variation in total personal $PM_{2.5}$ exposure.

Paper III presents total personal exposures, residential indoor, residential outdoor, workplace indoor and urban ambient air concentrations of NO_2 in three *EXPOLIS* centres (Basel, Switzerland; Helsinki, Finland and Prague, Czech Republic). Log-linear multiple regression and time-weighted deterministic models to predict total personal exposure were constructed using the microenvironment and ambient air concentrations, combined with information about various residential indoor and outdoor factors and sources, and time-activity data, which were collected using questionnaires. Data measured at ambient fixed site monitoring sites alone were a poor predictor for variation in total personal NO_2 exposure, but adding personal questionnaire information improved the predictive power.

Paper IV compares NO_x and NO_2 concentrations predicted using an urban dispersion modelling system with the results of an urban air quality monitoring network. A statistical analysis to determine the agreement between the predicted and measured hourly time series of concentrations at four permanently located and three mobile monitoring stations in the Helsinki Metropolitan Area in 1996-1997 was performed. Agreement between the measured and predicted datasets was found to be good. The difference between model predictions and measured data was also analysed in terms of meteorological parameters. The modelling system tended to underpredict the concentrations in convective atmospheric conditions and overpredict them in stable conditions.

Paper V describes a model developed to evaluate the average exposure of an urban population to ambient air pollution. The model combines the predicted concentrations, location of the population and time spent at home, at the workplace and at other places of activity. Utilization of the modelling system has been illustrated by showing selected numerical results of NO_2 in the Helsinki Metropolitan Area in March 1996. The computed results illustrate the temporal and spatial variations in concentration, population density and exposure to ambient origin air pollution.

6 Conclusions

Ambient fixed site $PM_{2.5}$ and NO_2 concentrations behave quite differently. Ambient $PM_{2.5}$ concentrations are evenly distributed in the Helsinki Metropolitan Area. Diurnal and temporal

variations in $PM_{2.5}$ are also quite moderate. In contrast, ambient NO_2 (as well as PM_{10}) concentrations have clear diurnal, temporal and spatial variability.

Ambient fixed site $PM_{2.5}$ explained 48% of the variation in total personal (cross-sectional, short-term) exposure during the night. However, in the daytime, ambient fixed site $PM_{2.5}$ could explain only 15% of total personal exposure variation. Therefore, $PM_{2.5}$ data from ambient fixed site monitors adequately explain the total short-term $PM_{2.5}$ exposure of less active population, such as retirees, but poorly explain the exposure of the active working-aged population. Ambient fixed site NO_2 explained only 19% of the total personal (cross-sectional, short-term) exposure variation when data from Helsinki, Basel and Prague were combined (11% for Helsinki data only). Thus, NO_2 data from the ambient fixed site monitor alone are a poor predictor for variation in total personal (cross-sectional, short-term) NO_2 exposure variation.

Adding personal questionnaire information can, however, significantly improve predictive power of the concentrations measured at the ambient fixed site. For instance, log-linear regression models using ambient fixed site monitoring data combined with residential and work characteristics explained 37% of the variation in total personal NO_2 exposure (32% for Helsinki data only).

The highest predictive power to estimate total personal NO_2 exposures belonged to the regression models, which were based on measured microenvironment (residential indoor and outdoor and workplace) concentrations. They predicted 75% of variation in total personal NO_2 exposure both for Helsinki alone and for all centres combined.

The time-weighted microenvironment exposure model, which required time-activity data, predicted almost as well as the best regression models did, i.e. it explained over 74% of the total personal exposure variation (all centres combined).

Our exposure model was the first step in evaluating the average exposure of urban populations to ambient air pollution, and to illustrate these results using the Geographic Information System (GIS). It looked quite promising when compared with measured data. This kind of modelling enables the evaluation of not only current but also past and future scenarios. This model will be used to evaluate the impacts of different scenarios in the latest revision of the Transport System Plan in the Helsinki Metropolitan Area.

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