EU contracts ENV4-CT96-0202 (five centres) and ERB IC20-CT96-0061 (Prague)

Final Report:

Air Pollution Exposure in European Cities: the *EXPOLIS* Study

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Final report /Air Pollution Exposure in European Cities: the EXPOLIS Study

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0. ABSTRACT

Epidemiological literature of the 1990=s has revealed surprisingly large public health impacts associated with present common air pollution levels in North American and European cities. Any causal explanation of the health effects of air pollutants must go through exposure, yet, prior to *EXPOLIS* no large, population based air pollution exposure studies have been conducted in Europe, and consequently no European database of air pollution exposures of urban populations has existed until now. *EXPOLIS* is a European multicentre study for measurement of air pollution exposures of working age urban populations. The selected urban areas are Athens, Basel, Grenoble, Helsinki, Milan and Prague. The main objectives of *EXPOLIS* are:

* To assess the exposures of European urban populations to major air pollutants.
* To analyse the personal and environmental determinants and interrelationships to these exposures.
* To develop an European database for simulation of air pollution exposures.

These objectives were pursued by measuring the personal exposures, home indoor and outdoor and workplace levels of $PM_{2.5}$, VOCs and CO of approximately 500 subjects representing the adult populations of the selected cities.

The field work continued from summer of 1996 to winter of 1997-98. Identical sampling equipment, operating procedures, time-microenvironment-activity diaries, questionnaires, database and data entry tools were used in each Centre. To assure comparability of the data from the 6 cities in 6 countries, a strict QA/QC protocol was established and the field work was supervised by the QA Unit of KTL. Standard operating procedures were prepared for all subject, laboratory and field procedures, and the *EXPOLIS* field teams were trained in four joint workshops. VOC laboratory analyses were intercalibrated by the European Commission / Joint Research Centre (EC/JRC) Environment Institute in Ispra. Other techniques were intercalibrated between the teams.

This paper describes the main design features of the European Union 4^{th} Framework RTD Programme funded multicentre study; Air Pollution Exposure Distributions of Adult Urban Populations in Europe (*EXPOLIS*). The *EXPOLIS* Centres are KTL- (coordinating Centre) in Helsinki, University of Athens, University of Basel, University Joseph Fourier in Grenoble, University of Milan, Regional Institute of Hygiene of Central Bohemia in Prague, VTT in Helsinki, and RIVM in Bilthoven. More detailed descriptions of the materials, methods, results and conclusions of this large, multiCentre and multidimensional study will be published later in more focussed articles.

1. INTRODUCTION

Why Air Pollution Exposure?

Measuring of the outdoor air levels and trends of pollutants at fixed ambient air quality monitoring sites together with modelling outdoor air concentrations with a multitude of dispersion models has been the traditional way of evaluating urban air quality and estimating the needs and effectiveness of air pollution abatement programmes. The possibility/potential of harmful health effects of air pollution has been estimated by comparing these levels to air quality guideline values. This logic has been challenged by a number of recent developments in both air pollution and scientific knowledge.

To keep the maximum air pollution levels at ground level air below the guideline values, industrial and power plants were in the 1960's and -70's equipped with increasingly higher stacks, and the rapidly growing road traffic was directed from the city streets to wider highways further away from the housing areas. These policies, based on the philosophy "*solution to pollution is dilution*" together with the growing traffic, industrial production, and energy demand greatly expanded the areas affected by air pollution. Yet, the maximum local and short term pollution levels within those areas have mostly been reduced. In the later 1970's and beginning of the -80's, flue gas desulphurisation together with increasing replacement of coal with natural gas began to reduce the SO_2 emissions, improving combustion technologies in heat generating stations began to reduce NO_x emissions, and towards the end of the 1980's catalytic converters (in Europe, 10 years earlier in the U.S. and Japan) began to slow down the increase of traffic generated CO, VOC, and NO_x emissions.

Fifteen-twenty years ago it started to become evident that because people spend 80-95% of their time indoors, human exposure to air pollution is dominated by indoor air pollution, which is partly outdoor air pollution that has penetrated indoors and partly pollution from indoor sources. Indoor spaces, where people are exposed, consist of millions of semi-closed microenvironments, offices, homes, rooms, kitchens, industrial workplaces, shops, restaurants and the like. Outdoor microenvironments, such as street canyons, highways, filling stations and even home gardens have also been found to be important for certain exposures. In addition to microenvironments, also activities, such as garden work with petrol driven lawn mowers or pesticide sprays, cooking with gas stoves, driving a car in traffic, or hobbies such as woodwork or painting, even ice hockey playing, are all important determinants of the human exposure to air pollutants.

Recent investigations by American epidemiologists, Dockery *et al.* (1992, 1993 and 1994), Schwartz *et al.* (1992) and Pope *et al.* (1995), re-analysis of the Six-Cities-Study data by the Health Effects Institute (HEI Oversight Committee, 1995)), and European multicentre projects such as APHEA (Katsouyanni *et al.* 1995, Dab *et al.* 1996, Katsouyanni *et al.* 1996, Ponce de Leon *et al.* 1996, Pönkä and Virtanen 1996, Schouten *et al.* 1996, Schwartz *et al.* 1996, Spix and Wichmann 1996, Sunyer *et al.* 1996, Touloumi *et al.* 1996, Vigotti *et al.* 1996, Zmirou *et al.* 1996), the Swiss studies on adults (SAPALDIA; Ackermann-Liebrich *et al.* 1997), and children (SCARPOL; Braun-Fahrländer *et al.* 1997) have radically changed our understanding of the health effects of air pollutants. Ten years ago, most experts would have agreed that severe health effects of the present air pollution levels in North America and Western Europe are rare. We now estimate that differences of air pollution levels, especially fine PM, in time and space are associated with tens of thousands of cases of respiratory and cardiovascular morbidity and mortality in Europe annually, and significant reduction in the length of life of large populations (WHO 1995).

However, although the mentioned time-series and cohort studies are based on ambient air data from urban air quality monitoring networks, the harmful health effects of urban air pollutants are not caused by the levels of air pollutants at those fixed monitoring sites, but instead by the personal exposures of the millions of individuals in their daily activities in indoor and outdoor urban environments and in

commuting between them. Such personal exposures may vary substantially between subgroups and individuals. Thus personal exposure data are an important prerequisite for risk assessment.

A number of air pollution studies where personal exposures have been monitored have been done, but rather few on representative population samples.
Annex I: Table 1 introduces the main design features of such already published exposure studies. Most personal exposure studies have been done on NO_2, because it is a significant air pollutant, has both outdoor and indoor sources, and can be easily monitored with cheap passive samplers (Hoek *et al.* 1984, Fischer *et al.* 1986, Quackenboss *et al.* 1986, Ryan *et al.* 1989, Özkaynak *et al.* 1993, Song *et al.* 1993, Xue *et al.* 1993, Spengler *et al.* 1994, and Alm *et al.* 1998). Personal exposures to ozone have been studied in two small scale studies in Switzerland and the Netherlands (Monn *et al.* 1993, Fischer *et al.* 1993). The Washington-Denver CO study covered one pollutant and two cities (Ackland *et al.* 1985, Jungers *et al.* 1985, Ott *et al.* 1988, Wallace *et al.* 1989, Mage *et al.* 1989). VOC exposures have been studied in one population based study in California (Hartwell *et al.* 1987), and in another large indoor air and exposure study in Germany (Hoffmann *et al.* 1996). Nicotine as an indicator of passive tobacco smoke exposure has been monitored with passive personal samplers on a random sample of American non-smoking women (O'Connor *et al.* 1993). Lioy et al (1990) were the first to collect personal PM_{10} exposure samples. The Particle-TEAM study collected both PM_{10} and nicotine exposures of residents of Riverside, CA (Wallace *et al.* 1993, Thomas *et al.* 1993, Clayton *et al.* 1993, zkaynak *et al.* 1996). In a Dutch study on personal PM10 and fine PM exposures were measured from 50-70 year old adults and schoolchildren (Jansen *et al.* 1997, 1998) . Personal exposures to PAH were studied by Waldman *et al.* (1987) and both PAH and organic mutagens were analysed in the Czech-U.S.EPA health study in the Teplice area (Watts *et al.* 1994). Reported multicomponent exposure studies are few. The LIILA study in Helsinki is the only one with personal exposure sampling of preschool children, and multicomponent gaseous (CO and NO_2) exposures (Alm *et al.* 1994, Alm *et al.* 1998). The daily personal exposure to PM_{10}, NO_2 , CO, Benzene, Toluene and TVOCs have been studied in 100 office workers living in the metropolitan area of Milan (Carrer *et al.* 1997). In addition there have been a few studies where personal exposures to multiple air pollutants have been monitored in traffic situations (Bevan *et al.* 1991, Wijnen *et al.* 1995).

Most of these data are American, or collected from non-representative and often small numbers of subjects. Clearly missing have been European representative and comparable air pollution exposure data, which could be used to assess air pollution exposure distributions in populations, to search for the factors that are associated with high exposures or to evaluate exposure distributions within specific subpopulations.

Suggested Research in Europe

The MRC Institute of Environment and Health (Leicester, U.K.) in collaboration with the WHO Centre for Environment and Health (Bilthoven, The Netherlands) organized a European **Workshop on Air Pollution and Health "Understanding the Uncertainties"** for 50 invited international experts on 2-4 February, 1994, in Leicester, U.K. One of the research topics that this workshop suggested was this:

"Personal activity patterns and variability within and between countries"

The discussion of this area of uncertainty led the working group to propose a four stage study, which could be used to evaluate the personal exposure of the European population to air pollutants as follows:

i. Firstly instrument development for personal monitoring of some pollutants is necessary, e.g. small portable continuous analysers for PM_{10}.

ii. Small scale detailed studies of personal exposure should be undertaken. This would include personal sampling, monitoring of microenvironments, and assessment of activity patterns in different settings. Sensitive groups would be studied as a priority.
iii. The small scale studies described in (ii) above would be followed by a Europe wide survey of relevant activity patterns.
iv. Finally, Europe wide population exposure distributions could be modelled (using Monte Carlo techniques).

- The outcome of the four stage study programme described above could ultimately allow the effectiveness of control measures to be predicted both in terms of cost effectiveness and the effectiveness of risk management strategies.

- Similarly the health impact of changes in the environment from future developments could be predicted.

- The data generated would also be useful for planning epidemiological studies and assessing the value of fixed point measurements in assessing personal exposures.

This study attempts to fulfill the strategy level (ii). The advantages of such a study are those listed above.

ECA: Air Pollution Epidemiology

In 1989 the principal investigator of EXPOLIS, was selected by Commission of European Union, DG XII to coordinate a new European Concerted Action on Air Pollution Epidemiology. This programme has up to now produced methodological reports; Exposure Assessment (Williams (ed.) *et al.* 1992), Health Effects Assessment, and Study Designs in Air Pollution Epidemiology (Katsouyanni (ed.) *et al.* 1993), two regional reports; CEC-East European Workshop on Air Pollution Epidemiology (Budapest, May 22-25, 1991)(Rudnai (ed.) 1992), and Air Pollution and Health in the Mediterranean Countries of Europe (Athens, October 8-10, 1992)(Katsouyanni (ed.) 1993). New reports are in progress on Health Risk Assessment of Air Pollutants, Time Activity Patterns (Workshop in Basel, February 14, 1994), Workshop on Air Pollution Epidemiology - Experiences in East and West Europe (Berlin, November 14-15, 1994), Socioeconomic and Cultural Factors in Air Pollution Epidemiology (Workshop in Brussels, March 21-22, 1995).
In addition to these workshops and methodological and regional reports, the ECA Air Pollution Epidemiology Programme was the birthplace of a number of EC 3rd and 4th Framework Programme funded European multicentre studies on the effects of air pollution on health. The studies that relate to the risks of air pollution and health may be viewed according to their coverage of the *emission* → *ambient air pollution* → *indoor air pollution* → *exposure* → *dose* → *health* chain. A full risk assessment covers the whole range.

Pollution Effects on Asthmatic Children in Europe (PEACE), (*ambient air pollution* → ... → *health*) was a panel study design that combined the efforts of 14 centres, all working with the same protocol, to investigate the European urban-rural, south-north dimension of air pollutants and the short term effects of low levels of respirable particles (PM10) and NO2 on the incidence of respiratory symptoms in asthmatic schoolchildren. PEACE was coordinated by Professor Bert Brunekreef from the University of Wageningen, Holland. The PEACE I study is now finished and mostly published. PEACE II is based on the elemental analyses of the PM samples collected in PEACE I and this phase is still ongoing. (1993 -)

The second multi-Centre study, **Short Term Effects of Air Pollution on Health: An European Approach Using Epidemiologic Time-Series Data (APHEA)** (*ambient air pollution* → ... → *health*)

was a time series study that uses death registers and hospital records from 12 major European cities to investigate the health effects of urban air pollutants. APHEA is coordinated by Professor Klea Katsouyanni from the University of Athens. Its aim is *exposure* → *health* relationship assessment, although ambient air pollution is used as a proxy for exposure. Within the framework of the project, the methodology of analysing time series data, as well as that of performing meta-analyses, are further developed. APHEA II focuses on the issues of the roles of individual pollutants and their mixes, dose response shape, and the possible role of harvesting in the observed daily pollution - mortality associations. (1993 -)

PHARE (DG I) Project on Environmental Health and Air Pollution (CESAR) (*ambient air pollution* → ... → *health*) was funded by the CEC and World Bank, and coordinated by Dr. Erik Lebret from RIVM, Prof. Bert Brunekreef from the University of Wageningen, and Dr. Tony Fletcher from the London School of Hygiene and Tropical Medicine. It focussed not only on the links between air quality and health, but also on the promotion of coherent epidemiological study designs and methodologies in the six PHARE countries (Poland, the Czech and Slovak Republics, Hungary, Romania and Bulgaria) and was divided into 3 subprograms: 1) on air pollution and respiratory diseases of children, 2) on quality assurance where a workshop and interlaboratory comparisons have been conducted on air pollution measurements and epidemiological methods, and 3) on a risk perception and communication survey. (1994 -1996)

Analysis of Small Area Variation in Air Quality and Health: A Methodological Study (SAVIAH) (*ambient air pollution* → ... → *exposure* → ... → *health*) applied, tested and evaluated new and emerging methodologies in the field of epidemiology, geography and pollution. This study combined the efforts of 8 centres in The U.K., The Netherlands, Poland and The Czech Republic and was coordinated by Dr. Paul Elliot at the London School of Hygiene and Tropical Medicine. The study aimed at 1) a questionnaire survey among parents of 5000 children, 7-11 years of age, 2) a series of air pollution surveys for NO_2 and SO_2 using passive samplers, 3) geographical information systems for the study areas, and 4) methods development for examination of relationships between health, air pollution, socio-economic and other data. (1993 - 1996)

The following studies were started in the 4th framework programme mostly in 1996, and only some initial results from them are available in 1998.

AULIS (ongoing) concentrates on the *exposure* → *dose* step to evaluate how sensitive and specific different biomarker techniques are for air pollution exposure assessment. It is the first European scale biomarker study with a sufficient population sample based on power estimation.

CEPLACA (ongoing) covers broadly the *emission* → *ambient air pollution* → *exposure* → *dose* chain, including terrestial and aquatic bioaccumulation, but focuses on the narrow issue of Pt, Pa and Rh from auto catalysts.

EXPOLIS (ongoing) investigates the *ambient air pollution* → *indoor air pollution* → *exposure* chain in European cities with an objective to produce and validate tools for predicting exposure consequences of personal behaviour and urban development alternatives.

ULTRA I-II (ongoing) covers *ambient air pollution* → *exposure* → *health*, and focuses on the means and objectives of fine particulate monitoring, relations between ambient air levels and exposures, and cardiovascular consequences.

TRAPCA (starting) aims at *emission* → *ambient air pollution* → *exposure* assessment (by modelling and measurement) of small children to traffic air pollutants - differential exposure being viewed as a proxy for differential risk.

As most of these studies are still ongoing, most of their conclusions and societal impacts lie still ahead. Some interesting conclusions can be made already:

- The broad time patterns of air pollution are often similar over large areas of Europe, and the differences in their levels between urban and rural sites may be quite small. The most significant European air pollution gradient is North-South, not West-East (PEACE-I and CESAR).

- The day to day variation of urban air pollutants at the present levels in European cities is associated with significant short term variation in cardiovascular and respiratory diseases and death (APHEA-I).

- Different optical and electrical fine PM counting methods agree well for < 0.5 μm, but less for larger particles (ULTRA-I).

- Many potential biomarkers of air pollutants appear to be too unspecific to be useful as biomarkers of specific exposures (AULIS).

- The concentrations of Pt, Pa and Rh in urine samples of urban children are low and their ratios in urine are different from their respective ratios in auto catalysts (CEPLACA).

- The fine PM exposures are dominated by smoking and outdoor air quality. Low socioeconomic status increases workday exposure, and young age night time exposure. Two groups of closely intercorrelated VOCs dominate the total VOC exposure (EXPOLIS).

1.1. Personal Air Pollution Exposure

1.1.1. Definition of exposure

Exposure of an individual to a pollutant can be defined as the contact concentration of the pollutant experienced by the individual (Georgopoulos and Lioy 1994), or as a coexistence of an individual and a pollutant in the same microenvironment (Ott 1993). Thus the exposure relates directly to the pollutant of interest, to the individual and to the timing and duration of exposure.

Air pollution levels show substantial temporal and spatial variation. This can be taken into account by the concept of personal integrated exposure over time period $t(t_0, t_1)$ for individual i:

$$E_i = \int_{t_1}^{t_2} c(t)\, dt \qquad (1)$$

where c(t) is the instantaneous concentration of the pollutant of interest at time t.

1.1.2. The time response of the personal exposure

The full data of personal exposure to certain pollutant can be expressed only as the full time series of instantaneous exposure (concentration) values experienced by the individual.

Depending on the exposure assessment needs, the personal exposure data can and must be reduced. This should be done in a way that preserves all the data relevant for the occurrence of health effects.

Personal exposure can be integrated for different time periods $t(t_0, t_1)$. The length of the time period should [ideally] be selected based on the physicokinetic mechanisms of corresponding health effects. Integrating (or averaging) time should be defined "backwards", starting from considerations of the time scales involved in the processes of the dose/response component of the exposure system (Georgopoulos and Lioy 1994).

This integrating or averaging functions as a low pass filter, removing all higher frequency concentration variation components from the data, thus reducing the data. If the low pass cutting frequency is selected correctly, no health relevant data is lost.

1.1.3. Time response of adverse health effects

The biological effects of exposures to toxicants result from its dose in a sensitive target organ. The actual level of toxicant concentration in the target organ depends on the exposure of the individual, toxicokinetics and metabolism of the substance in the organism, and accumulation and removal functions in the target organ.

Let's assume there is a threshold concentration of the pollutant in the target organ in which an adverse biological effect will occur. There are also complex accumulating and removal functions (which depend on biological, personal etc. properties). Then, if the accumulated concentration in the target organ will stay below the threshold limit for the life time of the individual, there should be no health effect of the pollutant to the individual.

On the other hand, if the threshold concentration is exceeded, then the health effect may occur.

The highest concentration in the target organ is the function of the exposure time series for the whole accumulation time, as well as the accumulation and removal functions. Thus no shorter time period for assessing the exposure can be set than the time period during which the integrated difference of the accumulating function and the removal function exceed the threshold concentration. This time period can vary from the shortest relevant exposure period to the life time.

The shortest relevant exposure period is the time during which the accumulation function can raise the target tissue concentration from zero to the threshold limit, and it is (probably) a function of the exposure concentration (e.g. higher concentration yields shorter accumulation time).

Consequently two exposure threshold limits can be defined. The lower exposure level threshold limit is the highest level that will never lead to a target organ dose threshold. The higher exposure level threshold limit is the level that will increase the target organ dose from zero to threshold over the integration time. Consequently, the lower exposure level threshold limit is for each pollutant independent of exposure integration time, but the higher exposure level threshold limit is a function of the integration time for each pollutant.

1.1.4. Personal Exposure Monitoring

As soon as reasonably inexpensive integrating personal passive sampling devices (PSD) and continuously recording personal exposure monitors (PEM) became available in the 1970's and '80's, these devices have been used for direct measurement of personal exposures as well as measurement of air pollution levels in representative micro-environments (MEM). They have the advantage of providing direct and detailed information of personal exposures, if the measurements are correctly done with adequate techniques. The disadvantage of the PSD is that all short term exposure information is lost. The disadvantage of the PEM is the cost of the equipment, and the amount of work in calibration, application and data reduction. The disadvantage of the use of MEM data is that the selected microenvironments may not represent the most relevant exposures, and the microenvironmental concentrations may not be the same when people - with their activities - are present as when they are absent.

1.2. Population Air Pollution Exposure

The **full data of exposure** to certain pollutant can be expressed only as the **full time series of instantaneous exposure concentration values experienced by each individual in the population**. For almost any application these are much more data that can be collected at any reasonable cost or utilized in any analysis. For many air pollutants the individual differences between the threshold concentrations (susceptibilities depending on age, health, activity and genetics) as well as exposure scenarios (time series depending on indoor, occupational and personal activity) vary significantly, or no threshold concentrations are known. Consequently, an exposure assessment must be based on data which are very much reduced from the ideal of a full time series of the exposure of each individual in the population/sample. This data reduction should be done in a way that preserves all the data relevant for the risk assessment. The length of the exposure integrating/averaging time, Δt, should [ideally] be selected based on the physicokinetic mechanisms of corresponding health effects. Δt should be defined "backwards", starting from considerations of the dose/response component of the exposure system (Georgopoulos and Lioy 1994). The data reduction can proceed over three orders. The following reductions assume that the averaging time of interest is 24 h, but, of course, it can also be e.g. 1 h or 1 year.

An example to describe the consecutive steps in reducing the full data set of personal exposures of a population sample is the THEES study of the 24 h personal PM_{10} exposures covering 14 days and 14 individuals in Phillipsburg, NJ, USA (Lioy *et al.* 1995). The full data set is presented in Figure 1.2.-1.

The **first order** data reduction combines all personal exposures for each day into daily frequency distributions. This is the **time series of the frequency distributions of personal exposures**. It preserves the time series data of the population, but all personal time series data are lost, thus losing the data that relate to longer or shorter term individual health effects.

Such a database can be used for identifying the days in which given exposure limits are exceeded within the population, and the daily percentages of the population exceeding such limits. For air pollutants, which do not have significant indoor sources, the time series of ambient air pollution levels measured at fixed air quality monitoring sites could be used as an approximation of the time series of the mean or median personal exposures, but the exposure frequency distributions around these daily means must be obtained from other information sources.

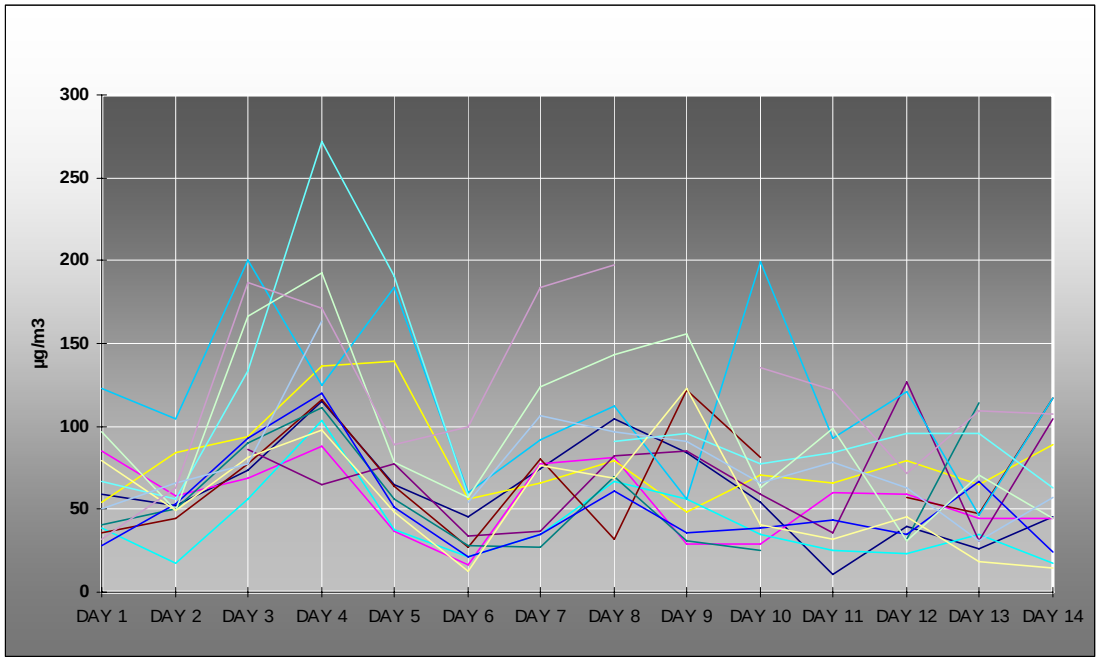

Figure 1.2.-1. Personal PM_{10} exposures (in Δ g/m^3) of 14 individuals in 14 days in Phillipsburg, NJ (Lioy *et al.* 1990).

A time series of frequency distributions of personal exposures in the THEES data is presented in Figure 1.2.-2.

The time series of the frequency distributions of personal exposures can be combined over time to form the **second order**, namely the **frequency distribution of the** (daily) **personal exposures**. Then, all time series data are lost.

Second order frequency distribution of personal exposures shows the percentages of the daily personal exposures in the whole population exceeding selected limits. It does not show, how these exceedances are distributed between different individuals and times. Because the data are considerably reduced, the frequency distribution of the personal exposures allows direct graphical and statistical comparisons of different exposure data sets, e.g. comparison of the exposures of the suburban with downtown residents, or residents of different cities. Data sets for frequency distributions of personal exposures have been produced in a number of air pollution exposure studies of representative population samples, however, in only three studies, THEES (Lioy *et al.* 1990), LiiLa (Alm *et al.* 1994, 1998), Jansen *et al.* (1998), have the exposures of the same individuals been measured repeatedly to allow for estimation of the first order time series of the frequency distributions of personal exposures.

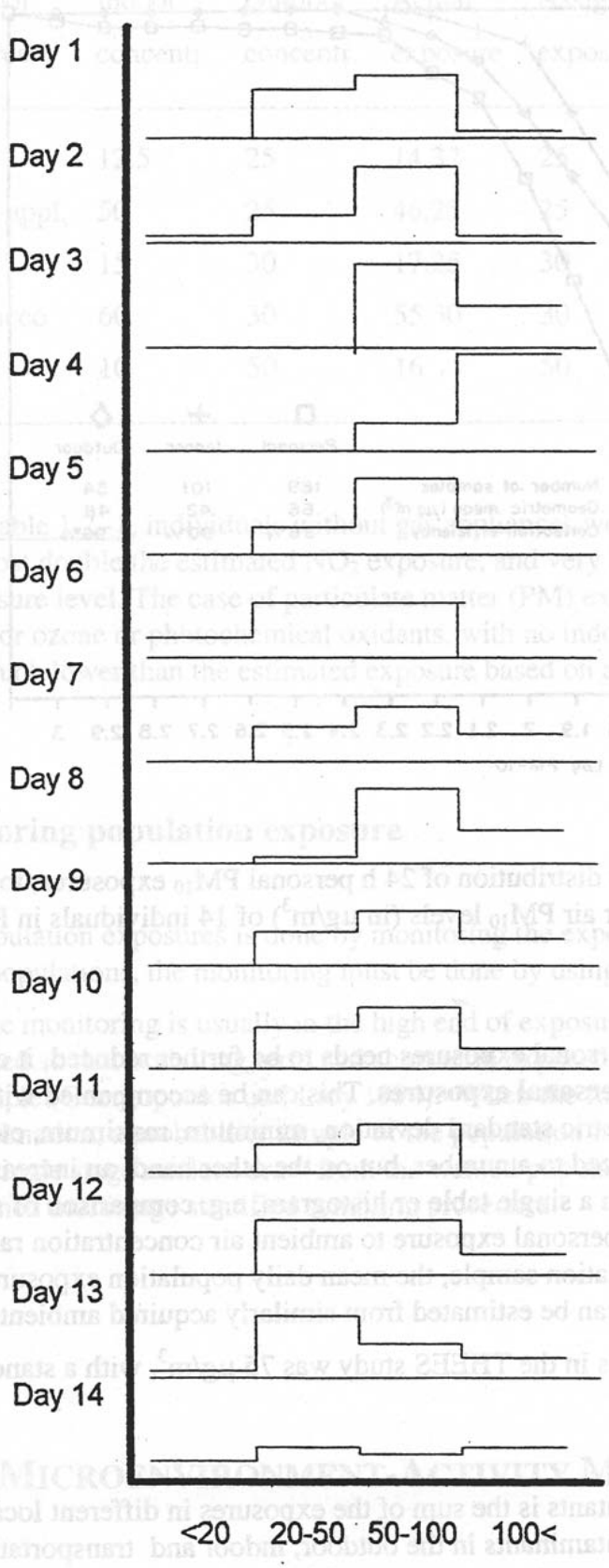

Figure 1.2.-2.

14 day time series of the frequency distributions of personal PM_{10} exposures (in $\mu g/m^3$) of 14 individuals in Phillipsburg, NJ (Lioy *et al.* 1990).

The second order frequency distribution of the personal exposures in the THEES data is presented in Figure 1.2.-3.

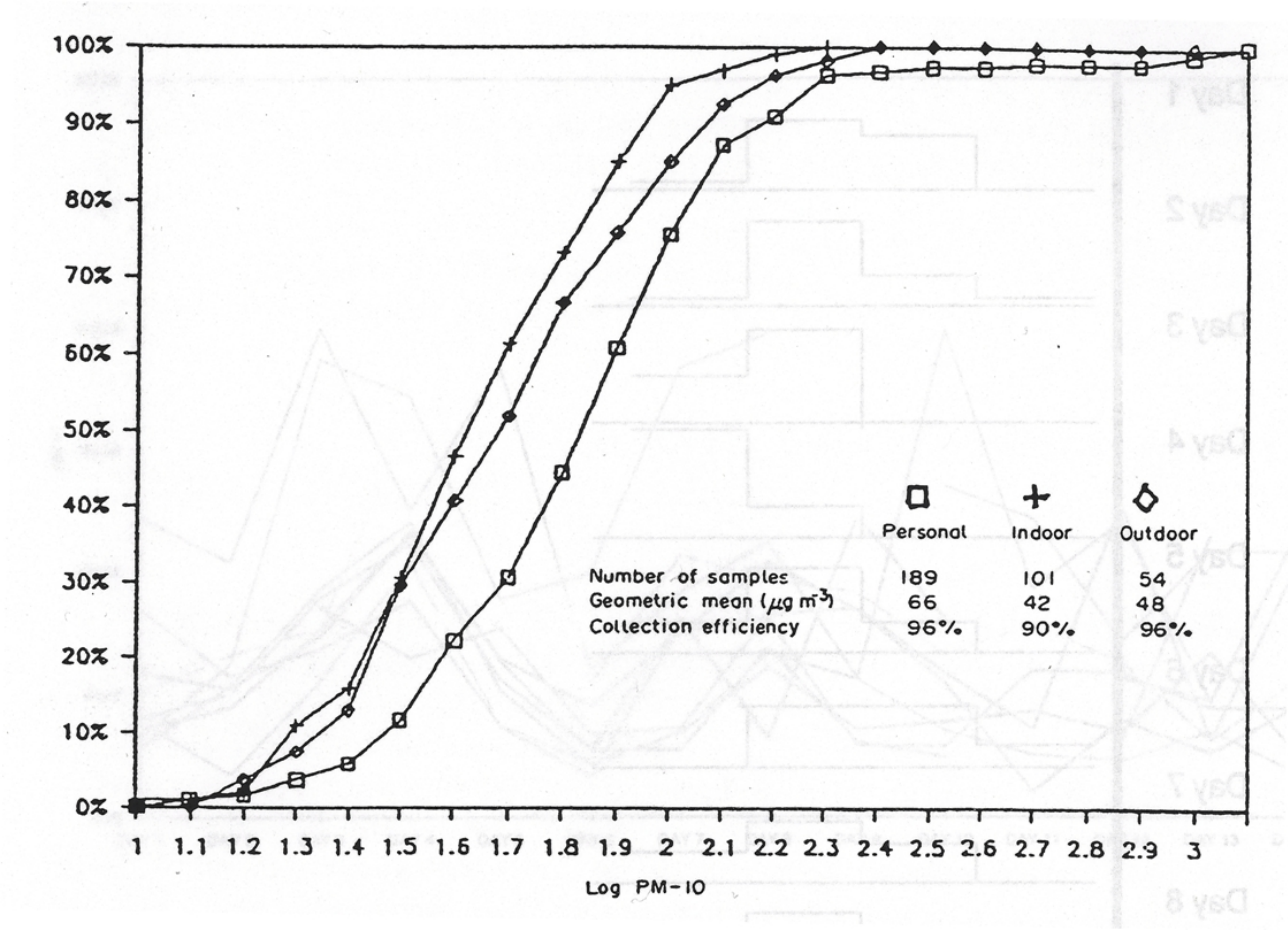

Figure 1.2.-3. Cumulative frequency distribution of 24 h personal PM_{10} exposures, together with ambient air and indoor air PM_{10} levels (in $\mu g/m^3$) of 14 individuals in Phillipsburg, NJ (Lioy *et al.* 1990).

If the frequency distribution of the personal exposures needs to be further reduced, it can be presented as the **mean or median of the** (daily) **personal exposures**. This can be accompanied with additional statistics, such as arithmetic or geometric standard deviation, minimum, maximum, etc. The frequency distribution data are now lost or reduced to a number, but on the other hand, an increased number of exposure data sets can be presented in a single table or histogram, e.g. comparison of several population subgroups in several cities. If mean personal exposure to ambient air concentration ratios have been determined for a representative population sample, the mean daily population exposures for a similar population in a similar environment can be estimated from similarly acquired ambient air quality data.

The mean of the daily PM_{10} exposures in the THEES study was 75 $\mu g/m^3$, with a standard deviation of 44 $\mu g/m^3$.

The total human exposure to air pollutants is the sum of the exposures in different locations and times. People are exposed to outdoor air contaminants in the outdoor, indoor and transportation environments, of which the indoor environment is the major component of the total exposure, because an overwhelming proportion of time is spent indoors. Table 1.2-1. presents, the range of misclassification which could result from using outdoor air concentration as an estimate for personal exposure. It is assumed that 85% of the time is spent indoors, 15% outdoors. The presence or absence of some common indoor sources is also considered.

Table 1.2.-1. Human exposure at home for different pollutants (Ackermann *et al.*, 1997)

Pollutants ($\mu g/m^3$)	Indoor source	Indoor concentr.	Outdoor concentr.	Actual exposure	Assigned exposure	Ratio actual/assigned
NO_2	---	12.5	25	14.37	25	0.57
NO_2	gas. apply.	50 25	46.25	25	1.85	
PM	---	15	30	17.25	30	0.57
PM	tobacco	60	30	55.50	30	1.85
O_3	---	10	50	16	50	0.32

According to Table 1.2.-1, individuals without gas appliances would have about half, with gas appliances almost double the estimated NO_2 exposure, and very few people would actually be at the estimated exposure level. The case of particulate matter (PM) exposures of smokers and non-smokers is quite similar. For ozone or photochemical oxidants, with no indoor sources, the actual exposure of everybody is much lower than the estimated exposure based on ambient air.

1.2.2. Monitoring population exposure

Monitoring population exposures is done by monitoring the exposures of the individuals separately. In case of larger populations, the monitoring must be done by using population samples.

The focus in the monitoring is usually in the high end of exposures. For such purposes the samples should be formed so that both long term and short term exposed subpopulations are well presented in the population sample (Georgopoulos and Lioy 1994). When the focus is on population exposure distribution estimation, a probability sample of the population is essential. This can be achieved locally with a simple single stage random draw from the wanted population or in larger geographical area by a carefully designed multistage stratified sampling procedure.

1.3. Time-Microenvironment-Activity Measurement

Air pollution exposure of an individual is a consequence of the levels of air pollutants in different micro- and macroenvironments and the presence and activities of this individual in them. While air pollution monitors are used to assess the former, time-microenvironment-activity monitoring techniques are used to assess the latter. Such data is usually collected by printed personally filled time-microenvironment- activity diaries (TMAD). However more advanced techniques are also becoming available (Jantunen 1995). Various levels of microenvironmental differentiation have been used (see Annex I: Table 2). In more recent studies this differentiation has, as a consequence of gained experience, generally been more coarse than in earlier studies. The ECA Air Pollution Epidemiology Programme has produced a whole report on Time Activity Patterns in Exposure Assessment (edited by Ackerman-Liebrich *et al.* 1995).

1.4. Exposure Survey Designs

The basic design fundamentals of previously published studies on personal exposure or microenvironmental air pollution levels are summarized in Annex I: Table 1. The table contains studies, where the air pollution levels have been measured and modelled, as well as studies on measured personal exposures and simulated exposure frequency distributions within large populations.

* Most of the studies are dealing with a single pollutant component, mostly NO_2 (10 studies) or CO (6 studies). The remaining studies focus on pollutant mixtures, such as ETS/nicotine (5 studies), CO/NO_2 (3 studies), VOCs (2 studies), particulate matter (2 studies), or other and more complicated sets of pollutants (7 studies).

* Most of the studies are based on short term data representing days to weeks of exposure. 10 of the studies attempt to cover the whole year or more.

* The list contains a number of studies, where the target population is poorly defined or does not represent major fractions of urban or national populations. Only 12 of the 34 studies are based on probability (random) samples of large populations.

* The population sample sizes are typically small, from 10 to less than 100 for descriptive studies that do not attempt to produce data representing large, general populations. Population samples from 175 up to about 700 have been selected to represent general urban populations or subpopulations, such as pre-school children in, Washington commuters, or people living in gas range homes in L.A. Even larger populations up to several thousand have been selected for questionnaire studies, mostly on ETS.

For designers of human exposure studies a WHO Guide (1991) recommends to begin with the following steps:

- Define the overall objectives of this study.
- Define the target population.
- Define the pollutants and routes of exposure.
- Define the information needed.

For selecting the population sample the WHO Guide (1991) stresses the importance of obtaining a true probability sample (which may be stratified for practical purposes) of the defined target population and achieving a satisfactory participation rate. Satisfactory means 75% or more, which, however is often not achieved due to the burdens that typical exposure studies impose in the participating individuals. As for the population sample size the WHO Guide states that for a human exposure study, the total sample should contain at least 50 individuals from the target population. Collecting exposure data from the population sample should contain monitoring their exposures and administering diaries for time-activity information and questionnaires about occupational classification, potential sources of pollutants, smoking, cooking and heating fuels, travel patterns, and demographics.

Finally the WHO Guide (1991) specifically warns about four types of tempting shortcuts:

- Failure to use a proper (i.e. probability) sampling procedure
- Failure to skip the pretest (pilot)
- Failure to follow up non-participants (accept low participation rate)
- Inadequate quality control.

1.5. The Measured Air Pollutants

1.5.1. Particulate Matter; TSP, RSP, PM_{10}, $PM_{3.5}$, $PM_{2.5}$

TSP is abbreviated from Total Suspended Particulates as collected by the standard high volume sampler, RSP is abbreviated from respirable suspended particles, usually referring to particles smaller than 3..4.5 μm in aerodynamic diameter, and mostly applied in industrial hygiene measurements. PM_{10}, $PM_{3.5}$ or $PM_{2.5}$ refer to particulate matter, where particles larger than 10, 3.5 or 2.5 μm in aerodynamic diameter have been separated out - usually by impactor or cyclone type preseparators.

Sources

Particulate matter (PM) in the air has two different origins.
Coarse particles (>1.0 ... 2.5 μm) are mostly produced outdoors by mechanical erosion in wind, traffic, and materials handling - and indoors by cleaning activities by resuspension of floor dust and handling of textiles. They contain mostly soil minerals, non-volatile organics and textile fibres. Much of the coarse PM settles rapidly out of the air, but is also easily resuspended. The average atmospheric lifetime of coarse PM is minutes to hours, and it can travel from metres to kilometres in the air (hundreds of kilometres for the smallest end of the range). Therefore coarse PM levels are highly variable. In the absence of open windows, coarse PM penetrates poorly from outdoor to indoor air, i.e. indoor air coarse PM is mostly suspended by indoor activities.

Fine particles (< 2.5...3.5 μm) are emitted directly into the outdoor air as carbonaceous soot and polyaromatic hydrocarbons (PAHs) by incomplete combustion processes such as diesel engines and wood burning and into indoor air by tobacco smoking, cooking and unvented kerosene heaters. They are also produced in the atmosphere by chemical reactions of gaseous sulfur dioxide from coal and oil burning, diesel engines and some metal ore smelting processes, nitrogen oxides from practically all combustion processes, most importantly road traffic, fossil fuels burning and some chemical industries, gaseous ammonia from farming and volatile organic compounds (VOCs). Fine PM contains mostly sulphates, nitrates, polyaromatic hydrocarbons, and elemental carbon (soot). The fine PM has very low settling velocity in air. It sticks to any surface that it happens to hit. The average atmospheric lifetimes of fine PM is long, days to weeks, and fine PM can travel thousands of kilometres. Fine PM penetrates effectively through most ventilation systems, and their levels in air can be fairly uniform over areas extending over hundreds of kilometres.

Exposure

Past Particulate Matter Exposure Studies:

On one hand the recent epidemiological findings about the public health impacts of atmospheric PM and on the other hand the tremendous costs involved in significant reduction of the present PM levels in most regions of the industrialized world lead to increasing demand for better information about;

- what chemical and physical characteristics of the PM are most significant for the health consequences observed,
- what environmental, microenvironmental and individual characteristics are most significant for personal PM exposures, and
- how much can the PM related health hazards be reduced by different control measures.

Personal exposure studies can produce answers to these questions.

In an early study on personal PM exposures of respirable PM, 37 volunteers in Watertown MA and Steubenville OH carried personal samplers and filled time activity/diaries 12 h at the time (Dockery and Spengler 1981). The main results of this study were that the 12 h mean personal PM exposure levels are in reasonably good agreement with the mean outdoor respirable particulate concentrations. This agreement could be only slightly improved by a time weighed (indoor, outdoor, smoking) model.

Sexton *et al.* (1984) assessed personal PM exposures of 48 volunteers in Waterbury Vermont. The volunteers carried personal sampling pumps and filled time activity/diaries every other day for two weeks, and their homes were also equipped with similar indoor and outdoor PM samplers. Their main findings were that outdoor particle levels were not an important determinant of personal exposure, and personal exposure levels were systematically higher than indoor air levels, which again were higher than outdoor air levels. Personal 24 h average PM exposure levels were modelled with a simple time weighed, 3 variable (intercept, exposure to smoke, work, in transit) model. Predicted exposure using this approach agreed well with measured values, explaining 51% of the variance in personal exposure.

A total of 97 nonsmoking volunteers in two rural Tennesee communities took part in the next personal PM exposure measurement and modelling study (Spengler *et al.* 1985). Personal samplers with a cyclone preseparator that passes 50% of 3.5 μm aerodynamic diameter particles and 0% of 10 μm particles were used. The volunteers carried the personal samplers, their homes were equipped with indoor samplers and outdoor air levels were monitored by centrally located samplers in each of the towns. A total of 249 personal, 266 indoor and 71 outdoor air samples are included in the analysis. The results show that personal exposure levels of non-smoke-exposed people are higher than outdoor air levels, and that personal exposures of smoke-exposed people are nearly twice as high as those of the non-smoke-exposed. A regression model that includes the variables outdoor air PM, smoke exposure, employment status, time at home, time at work, time travelling, time in public (spaces), other time, and indoor PM explained 64% of the variance in personal exposure.

Lioy *et al.* (1990) used a sharp cut 10 μm personal impactor together with a 4 l/min personal pump in Phillipsburg, NJ, to evaluate personal exposures of 14 non-smoking individuals, 8 indoor PM_{10} samplers to monitor indoor microenvironments, and 4 outdoor PM_{10} samplers to monitor outdoor microenvironments, as a part of the Total Human Environmental Exposure study (THEES). In this first PM_{10} study personal exposure levels were again higher than indoor and outdoor levels, but the latter two levels as well as their statistical dispersions were about the same. During the two winter stagnation episodes individual exposures and outdoor microenvironmental concentrations were strongly influenced by the outdoor PM_{10}.
The personal 4 l/min impactor sampler was also used in a much larger study to evaluate personal PM_{10} exposures of the population of Riverside, CA in the PTEAM study (Wallace *et al.* 1991, Wallace *et al.* 1993, Özkaynak *et al.* 1993, Thomas *et al.* 1993, Clayton *et al.* 1993, Özkaynak *et al.* 1996). A stratified probability sample of 178 people carried personal monitors 24 h at the time for two 12 h samples. The particle concentrations inside and outside of the home of each of the 178 participants were monitored with stationary PM_{10} and $PM_{2.5}$ monitors, and ambient air levels were monitored at fixed sites with high volume PM_{10} samplers. Both gravimetric and elemental analyses were done. Over 95 % of the scheduled 2900 samples were taken during the 48 days of field work and analysed with very few equipment failures. Following each of the two 12 h monitoring periods the participants answered an interviewer administered recall time/activity questionnaire. Daytime personal PM_{10} exposure levels, as well as the levels of nearly all particle bound elements were elevated relative to indoor and outdoor levels. Nighttime personal exposure levels were lower than outdoor but higher than indoor levels. Smoking, cooking, dusting and vacuuming were again found to be dominant sources for high indoor particle loads. Reentrainment of house dust through activities not recorded in the questionnaire could

also be a source of increased exposure. PM_{10} and $PM_{2.5}$ concentrations in smoking homes were considerably greater than those measured in non-smoking homes. Correlations of personal PM_{10} exposures with fixed site outdoor concentrations were low: 0.37 in the daytime and 0.54 at night. Modelling personal exposures with a microenvironmental model partially accounted for the excess personal exposure.

Kamens *et al.* (1991) looked at the particle size distributions, time variation and causes of the particle levels, and composition of the indoor air particles in three non smoking homes in Chapel Hill NC, over a three day period. They used indoor air samplers with 2.5 and 10 μm cut sizes, several electrical and optical aerosol analysers to obtain particle size distributions and short term particle level variations from 0.01 to 19.4 μm. The main findings were that as an average the particulate mass was nearly evenly distributed between the three aerodynamic particle size ranges, 37% in < 2.5μm, 26% between 2.5 and 10 μm, and 37% in > 10 μm. Aerosol size information from the automated instruments suggest that the most significant event for generating small particles was cooking, and vacuum sweeping was the most significant large particle generating event.

The main design features of the recent personal PM exposure studies can be found in Annex I: Table 1. Only one, the USEPA PTEAM study conducted in Riverside CA, is based on probability sampling from a defined population base, and can, thus be considered to produce an exposure estimate of a larger population than the sample itself.

Condensed Results of past Particulate Matter Exposure and Microenvironmental Studies:

The personal fine PM exposure levels and corresponding levels measured in microenvironments such as homes, workplaces, adjacent outdoor environments and central ambient air monitoring sites are presented in Annex I: Table 3. The observed (geometric) mean personal exposure levels for $PM_{2.5...3.5}$ have ranged 22 - 44 $\mu g/m^3$, home indoor 11 - 42 $\mu g/m^3$, home outdoor 10 - 38 $\mu g/m^3$, and central monitoring site 18 - 33 $\mu g/m^3$. The corresponding PM_{10} levels have naturally been higher, personal exposure 33 - 129 $\mu g/m^3$, home indoor 22 - 78 $\mu g/m^3$, home outdoor 18 - 83 $\mu g/m^3$, and central monitoring site 38 - 76 $\mu g/m^3$.

Annex I: Table 4 summarises the impacts of certain indoor air polluting activities on personal PM exposures and indoor concentrations. The most significant is, of course, smoking. A rough average $PM_{2.5...10}$ level increase in smoking v.s. non-smoking environments is 30 - 40 $\mu g/m^3$ or doubling of the non smoking level. In individual cases the impact depends strongly on the number of cigarettes smoked, size of the room or house and the ventilation efficiency. Cooking increases PM exposures 7 - 26 $\mu g/m^3$, unvented kerosene heaters 5 - 30 $\mu g/m^3$, wood stoves 0 - 10 $\mu g/m^3$, and ultrasonic humidifiers up to 542 $\mu g/m^3$. The humidifier case in both shocking and interesting, and probably representative to ultrasonic humidifier type only. The $PM_{2.5}$ level increase is nearly 20 times higher than smoking, and only by using distilled water can the level be reduced to same as smoking. These devices have been advertised as air cleaners (sic!).

Annex I: Table 5 presents source apportionments of personal, indoor air and ambient air $PM_{2.5 \ldots 10}$, except that no source apportionments have been published for personal PM exposures. Looking at indoor air data Table 5 shows again that where smoking takes place, it is responsible for 24 - 71 % of total PM mass. Cooking, where relevant, is responsible for about 25 % of indoor air PM. Wood burning is responsible for 3 - 21 % of PM, which comes mostly from outdoor air. Soil and road dust are responsible for 4 - 50 %, industrial and heating emissions for 10 - 38 %, and traffic emissions 5-30 % of indoor PM. The ambient air source apportionment is different, because the roles of smoking and cooking are very much reduced, and those of the other sources respectively increased. Most ambient air $PM_{2.5 \ldots 3.5}$ data is $\mu g/m^3$ and not % based: Secondary $SO_4^{=}$, NO_3^{-}, and NH_4^{+} particles form 11 - 22 $\mu g/m^3$ of the ambient air fine particulate mass, diesel vehicles contribute 4 - 12 $\mu g/m^3$, soil dust 1 - 23 $\mu g/m^3$,

industrial and heating emissions 4 - 6 μg/m^3, gasoline powered (cat) vehicles 1 - 5 μg/m^3, woodburning 1 - 4 μg/m^3, and cooking (not fire, but frying aerosols in LA) 2 - 3 μg/m^3.

Exposure-Response Assessment

Particles larger than 10 μm do not penetrate deep into the lung. Particles smaller than 2.5 μm do, about half of these particles are not exhaled, and, if insoluble, are only quite slowly removed from the lung tissues. Particles between 2.5 and 10 μm in aerodynamic diameter show intermediate behaviour that depends strongly on the breathing type (mouth or nose) and intensity (Bates *et al.* 1996).

Epidemiological Data:

Time Series Studies on Short Term Health Effects: Our knowledge about the health effects of the PM has improved considerably since extended time series of outdoor air quality data - mostly U.S. - from PM_{10} and $PM_{2.5}$ samplers has become available for epidemiological analyses. In a study on particulate air pollution and daily death rate in Steubenville OH (they analysed 15.000 deaths from 1974 to 1984), Schwartz and Dockery (1992) found a 6 % increase in daily deaths when daily TSP levels increased from 36 μg/m^3 to 209 μg/m^3. This result has later been confirmed in new time series studies in the U.S., China (Xu *et al.* 1994), and several studies in Europe, the largest being the collaborative APHEA study by Katsouyanni *et al.* (1995); in Lyon (Zmirou *et al.* 1996), Paris (Dab *et al.* 1996), Athens (Touloumi *et al.* 1996), Köln (Spix and Wichmann 1996), and Milan (Vigotti *et al.* 1996). Combined analysis of the APHEA data from 5 West European cities indicates a 2 % increase in daily deaths resulting from a 50 μg/m^3 increase in daily PM_{10} level (Katsouyanni *et al.* 1997).

Schwartz and Dockery have compared their results with those of previous studies, and conclude that there is a striking quantitative concurrence in the relative increase in total mortality versus particulates between different studies. The APHEA study has also produced disease and hospitalisation data (Anderson *et al.* 1997) which supports the findings of the mortality data, namely that existing levels of particulate air pollutants in West European cities have a significant impact on the cardiovascular and respiratory health of the urban populations.

Based on extensive review of the literature, WHO Air Quality Guidelines draft (6.10.1997) concludes that a daily outdoor air $PM_{2.5}$ increase of 25 μg/m^3 increases daily total mortality by 15% and total hospital admissions by 12.5 %, and that a daily outdoor air PM_{10} increase of 50 μg/m^3 increases total mortality by 15 (4 %), hospital admissions by 4.2 (1.6 %), cough by 23 % and asthma medication need by 17 %.
Time series studies leave one significant question open, namely, do air pollutants just synchronise inevitable deaths to a small extent (harvesting), or do they also significantly reduce the life expectancies of affected individuals and populations?
Studies on Long Term Health Effects: In the first cohort study on the relationship between annual average pollution levels and adjusted mortality-rate ratios in a cohort of 8.000 adults in six cities followed over 14-16 years, Dockery *et al.* (1993) found, after controlling for gender, age, smoking, education level, and occupational exposure, that all pollutant levels (TSP, $PM_{2.5}$, particulate sulphate, aerosol acidity, and SO_2) with the exception of O_3 were associated with increasing mortality. However, the association was strongest for $PM_{2.5}$. An increase in the annual average level of $PM_{2.5}$ from 10 to 30 μg/m^3 was associated with an increase of total mortality by 26 %, lung and heart disease mortality by 37 %, but all other causes of death were unrelated to air pollution.

In a larger cohort study on the associations between the $PM_{2.5}$ levels and adjusted mortality rate (50 cities, 295.000 individuals), Pope *et al.* (1995) find that an increase of the annual average $PM_{2.5}$ by 24.5 μg/m^3 is associated with a 17 % increase in total mortality and 31 % increase in lung and heart disease mortality.

In a U.S. study on the association of the death rate among 3.8 million babies 1..12 months of age with outdoor air PM_{10} levels during the first 2 months after birth shows that compared to the low exposure group ($PM_{10} < 31.8\ \mu g/m^3$), in the high exposure group ($PM_{10} > 45.5\ \mu g/m^3$) 10 % more babies died, 26 % more from sudden infant death syndrome, and 40 % more from respiratory causes (Woodruff *et al.* 1997).

Concluding from the three cohort studies, typical urban outdoor air levels of PM_{10} and $PM_{2.5}$ appear to increase long term death rate, i.e. reduce life expectancy. This increase is consistent between different studies, and seems to affect at least babies and adults. These observed increases in the long term death rates cannot be explained by acute effects of air pol
lution, but rather relate to increased morbidity that results in earlier deaths
This is also confirmed by studies which address long-term effects of air pollution on morbidity or physiologic measures such as lung function which strongly predict survival. In the Seventh Day Adventists Cohort Study, the incidence of chronic bronchitis increased with long-term ambient particulate levels (Abbey *et al.*, 1991). In the cross-sectional semi-individual (Künzli and Tager,1997) studies SAPALDIA and SCARPOL, lung function and morbidity of adults and children correlated with the long-term average ambient particulate exposure levels (Ackermann-Liebrich *et al.*,1997), (Schindler *et al.*,1998), (Braun-Fahrländer, *et al.*,1997).

The recent studies of Dockery, Schwartz, Pope and others have led to serious discussion about the needs to considerably reduce the levels of particulate air pollutants in urban air, and to revise the particulate ambient air quality standards (Friedlander and Lippmann 1994).

Toxicological Data:

Currently understood toxic mechanisms of individual harmful compounds or their combinations in the PM can hardly explain the observed mortality increases. The total mass of $PM_{2.5}$ particles inhaled into the lung during a full year, assuming 30 $\mu g/m^3$, is in the order of 1 mg. Indeed, this fact indicates that if the observed health effects of the atmospheric PM are real they may not depend on the specific chemical components of the PM. However, there are new, yet unpublished data on animal tests, which support the epidemiological findings (Godleski *et al.* 1997). Healthy dogs and compromised dogs with induced bronchitis and induced coronary heart disease, have been exposed to relatively clean urban air, in which the fine PM fraction has been concentrated by an order of magnitude (by a series of virtual impactors). Healthy dogs were not harmed, but when dogs with a cardiovascular or/and bronchial precondition were exposed similarly, significant short term mortality was observed.

Assuming that the considerable fraction of urban dwellers, who live with asthma, chronic bronchitis or coronary heart disease, do not differ significantly from these dogs, the test results indicate that the safety margin in the present day urban air fine particle levels and air quality guidelines is very small or nonexistent. The epidemiological studies point to exactly same conclusion.

A hypothesis by Seaton *et al.* (1995) suggests that ultrafine particles [mostly generated by atmospheric chemistry and physics of gaseous and vapour phase pollutants] are able to provoke alveolar inflamation, with release of mediators capable, in susceptible individuals, of causing exacerbations of lung disease and of increasing blood coagulability. The authors suggest that this hypothesis be tested epidemiologically and with animal experiments.

Indoor Air Particles?

All the available epidemiological data is based on relating PM levels measured at fixed urban outdoor air monitoring sites to long or short term mortality and morbidity data of populations or cohorts. How

do these findings relate to the health risks of indoor air particles? Individual exposures to PM (see also Annex I: Tables 3., 4. and 5.) can be divided to e.g. outdoor air PM (measured by most urban monitoring sites), outdoor microenvironmental or near field PM (most importantly traffic exhaust particles on busy streets), outdoor air particles that have penetrated indoors (ventilation air, open doors and windows, and air leaks), particles generated by indoor activities (cooking, heating, environmental tobacco smoke, etc.), and intentional personal exposures (smoking). In average, non-smoking individuals appear to acquire roughly one half of their PM exposures from outdoor air particles - note, mostly in indoor environments - and the other half from indoor and personal sources.

The near field and personal PM sources have typically a much stronger immediate impact on personal PM exposure levels than the outdoor air PM levels. However, outdoor air PM, especially the long lived and effectively penetrating fine PM, forms the large scale background on which the impacts of the near field, indoor and personal PM sources are added on. It has been argued, as a justification to the time series studies, and also shown (Jansen *et al.* 1998) that although the outdoor air PM level may be a poor predictor of personal PM exposure, the day to day difference in outdoor PM level is a better predictor of the day to day difference of the population (or group) PM exposure.

The question - can the risk estimates, based on the statistical association of population mortality and morbidity with outdoor air PM levels, be used to estimate the health risks of exposure to PM indoors - remains still unanswered. If the health effects of fine PM are mostly independent of the origin/composition of the particles, as some epidemiological studies seem to indicate, then indeed the risks of the indoor fine PM should be assessable on the basis of the outdoor air PM based epidemiological studies. If the health effects of fine PM do depend on their origin/composition, as toxicological plausibility seems to require, then the health effects of indoor PM cannot be directly assessed on the basis of the outdoor air PM based epidemiological studies. This problem, however, is lager in principle than in the praxis: Most of the individuals that are affected by elevated outdoor air PM levels are anyway exposed indoors. In the absence of smoking, outdoor air provides 50...60% of the indoor air particles.

Conclusions of Health Effects:

There are sufficient reasons to assume that the fine PM in urban outdoor (and indoor) air is hazardous to public health even at the presently common relatively low concentrations. We do not know yet (i) what characteristics make the particles harmful, although combustion generated particles are the most suspected, (ii) what characteristics make individuals more susceptible, although individuals compromised by cardiovascular or respiratory diseases are the most likely targets, or (iii) what biological mechanisms are responsible for the observed acute and long term morbidity and mortality increase, although inflammation is the most suspected.
The U.S. cohort studies on long term health effects suggest that a 10 $\mu g/m^3$ increase in the long term mean $PM_{2.5}$ level increases total death rate by 7-13 %. U.S.EPA (1996) concludes in its new Air Quality Criteria for PM that (very much abbreviated by the author);

- There is very much evidence that daily outdoor air fine PM ($PM_{2.5}$) is significantly increasing daily deaths and cases of disease at present concentrations (in North American cities).

- There is less, but still convincing evidence, that fine PM also reduces life expectancy.

- There is no clear evidence about what physical or chemical fine PM characteristics make it hazardous.

- There is no indication of a threshold level, below which fine PM is harmless (if one exists, it is below today's cleanest cities PM levels).

- Elderly individuals with cardiopulmonary diseases appear to be at highest risk, asthmatic children may also form a susceptible group.

All present epidemiological evidence is based on outdoor ambient air pollution data. The observed health risks of outdoor air PM result mostly from exposure to these particles in indoor environments (by necessity - people spend 90% or more of their time indoors. However, although the risks of fine PM from indoor sources (except smoking) are not known, this does not much affect risk assessment for indoor fine PM, because 50 - 65 % of it comes from outdoor air/sources. Therefore if we can accept an uncertainty factor of 2, we can apply the risk estimates from outdoor air PM to common indoor exposures as well.

The epidemiological studies do not support any threshold level, below which PM exposure could be considered safe to the general public. This is also the conclusion of a WHO expert group responsible for preparing material for the new WHO Air Quality Guidelines (Draft 6.10.1997). Instead of an air quality guideline value, the group suggests a unit risk for PM. The group concludes that:

- A 100 $\mu g/m^3$ increase in 24 h average PM_{10} exposure results in a 6...8 %, $PM_{2.5}$ exposure in a 12...19 % increase in daily deaths within a population,

- a 50 $\mu g/m^3$ increase in 24 h average PM_{10} exposure results in a 3...6 %, $PM_{2.5}$ exposure in about 25 % increase in total hospital admissions, and

- among asthmatics a 25 $\mu g/m^3$ increase in 24 h average PM_{10} exposure results in a 8 % increase in symptom exacerbation and bronchodilator use, and a 12 % increase in cough.

Even if a threshold level exists, it is likely to be so low that it is not relevant for outdoor or indoor air quality management in the urban areas where majority of the population lives. The epidemiological evidence and occupational hygiene experience suggest that the short term acute death risk of a 24 h PM_{10} level of 100 $\mu g/m^3$ is probably negligible for the healthy majority of the population, but amplified for babies in their first year(s) of life, and for the large numbers of, particularly elderly, individuals with and underlying cardiovascular or respiratory diseases.

A WHO, USEPA based research group has evaluated the health difference between two IPCC Climate Change scenarios, one Business as Usual, the other Effective Fossil Fuel Reduction. From year 2000 to 2020 the difference between the former vs the latter is 8.000.000 cases of excess deaths from higher fine PM from higher fossil fuel use alone (Working group 1997).

Sampling

PM_{10} and $PM_{2.5}$ can now be sampled by personal sampling pumps using a sharp cut impactor to separate the particles that are larger and smaller than 10 μm (or 2.5 μm) in aerodynamic diameter. These samplers can operate up to 15 h with a single set of batteries (Buckley *et al.* 1991, Thomas *et al.* 1994).

Reponen *et al.* (1995) and Mirme *et al.* (1995) have compared the measurements of black smoke (BS) as measured according to the OECD standard, and black carbon (BC) measured with an aethalometer (Magee Scientific) with size fractionated particle count data measured by the Tartu University developed Electrical Aerosol Spectrometer (12 size fractions between 0.01 um - 10 um). Both BS (r^2 = 0.91) and BC (r^2 = 0.93) data correlate very well with the $PM_{1.0}$ particle numbers (aerodynamic diameter between 0.01 and 1.0 μm). As the BS and BC analyses are not gravimetric but optical, they require only small samples, and can be considered as indirect but probably quite useful proxies for the $PM_{1.0}$ particle numbers.

Continuously measuring personal PM monitors do exist, but until recently they have not bee sufficiently sensitive for monitoring environmental particulate levels. There are, however, new instruments which are sufficiently sensitive for urban environmental monitoring, battery powered, relatively lightweight, and have memory capacity for hundreds of measurements. The new personal DataRAM monitor (MIE Inc. Bedford, MA), is based on passive air sampling and forward light scattering principle. It is sensitive for particles in the diameter range of 0.1 - 10 μm at concentrations down to 1 $\mu g/m^3$. The TSI DusTrack monitor (TSI, Inc. StPaul, MN) is also an optical aerosol monitor, but the analysed air flow is pumped at 2.5 L/min. Consequently it can be equipped with a size selective inlet (e.g. $PM_{2.5}$), but it is also bigger and has a much higher battery consumption. All optical aerosol analysers are sensitive to particle size distribution, the optical characteristics of the particles and atmospheric humidity, and need therefore be calibrated for each measuring condition and aerosol type. This greatly limits the applicability of optical aerosol monitors for personal exposure monitoring, where both the aerosol characteristics and the ambient air conditions change while the person moves from one microenvironment and activity to another.

1.5.2. Carbon Monoxide

Sources

CO is produced in incomplete combustion of carbonaceous fuels. In modern urban settings street traffic is the dominant source of CO. For indoor exposures gas stoves, gas fired water heaters and other unvented heating equipment may increase the indoor CO levels considerably from the outdoor levels - even to acutely dangerous levels. The atmospheric lifetime of CO is estimated to be in the order of tens of days, so within an urban airspace CO can taken as an inert, non reacting gas.

Exposure

The key design features of the CO exposure studies are summarized in
Annex I: Table 1.

Children:

Alm *et al.* (1994) studied personal CO exposures of preschool children in Helsinki, and in comparing exposure frequency distributions between different subgroups, found that the whole exposure frequency distributions were shifted upwards by gas stoves (which at that time in Helsinki burned man made town gas), parental smoking at home, and low socioeconomic status of the parents, that the highest 5 % of the exposure distribution was shifted upwards by commuting to and from the day care centre by car or bus vs walk or bike, and that location of home in the downtown vs suburban region produced no visible difference in the distribution curve. The 1 h personal exposures of the children showed no correlation with, but the lowest 8 h exposures were well predicted by the fixed monitoring data.

Adults:

Cortese and Spengler showed already in 1976 that the frequency distribution of fixed ambient air quality monitoring station CO data (Massachusetts) underestimate personal 1 h CO exposures during commuting by a factor of 1.4 - 2.1. No consistent relationship was observed between personal exposure during commuting and fixed station measurements over the entire range of values encountered. On the other hand, measurements at fixed stations were representative of 8 h population exposure. The personal exposures are strongly affected by individual modes of transportation. Individuals commuting by auto are exposed to twice as high daily maximum 1 h levels than people commuting by mass transit. Wind speed, wind direction, season and automobile age did not influence commuter exposure to CO.

The big CO exposure study that followed the early experiments was the USEPA Washington-Denver study. The methodology developed for this study combined statistical survey design techniques from social sciences with miniaturized automated PEMs in order to produce exposure profiles for a representative population sample. The resultant database was intended to be used for 1) assess the risk of a pollutant to public health, 2) interpret and understand data collected by conventional fixed air monitoring networks, 3) select fixed monitoring stations that better reflect population exposures, 4) allow investigators to better assess the impact of alternative regulatory strategies, and 5) permit investigators to develop models for predicting future exposure frequency distributions in response to changes in urban form, human activities, and population growth (Jungers *et al.* 1985).

The field work was performed in the winter of 1982-83, when 712 and 450 individuals, stratified probability samples from Washington, DC, and Denver, CO, carried personal CO-exposure monitors, each for one day. The results indicated again that while fixed monitoring station data underestimates maximum short term exposures, it represents rather well average population exposures. The personal parameters that significantly increased personal CO exposure were high occupational exposure and commuting to work (Ackland *et al.* 1985). In Washington, DC, the average CO exposure levels and times in commuting with automobile were 8.8 - 22.3 ppm and 34 - 69 min, in bus 3.7 - 10.2 ppm and 81 - 115 min, and in rail 2.2 - 5.2 ppm and 27 - 48 min. The highest exposure levels were recorded in garages. On street or highway the exposure levels decreased significantly with increasing automobile speed, 10 - 60 mph (Flashbart *et al.* 1987). At the lowest speed range, 10-30 km/h, this last finding seems to disagree with an earlier study of Rashidi and Massoudi (1980) from the streets of downtown Tehran, Iran. They found a sharp maximum street level CO concentration with a traffic speed of 30 km/h (19 mph). However, Flashbart *et al.* measured CO levels inside moving cars, Rashidi and Massoudi on the street. The American cars in 1983 were nearly all equipped with emission controls, while cars in Iran in 1976-77 were not. Also the urban traffic settings are quite different in downtown of Tehran when compared to the whole Washington, DC, Metropolitan Area.

The measured personal exposure levels, combined with each person's age, gender and height, were also used to model blood COHb levels (Coburn equation), and the modelled values were then compared to those determined from breath samples. The agreement was surprisingly poor. The model predicted as an average too low exhaled air CO levels, and the scatter of values was remarkable. (Wallace *et al.* 1988)

Because Denver, CO, and Washington, DC, have different climates, urban plans, and altitudes, the researchers have also studied the comparability of microenvironmental data collected from the two cities. The conclusion is that although many microenvironments in the two cities should have reasonably similar concentrations after correcting for ambient conditions, other microenvironments, such as automobiles in traffic (as expected due to different combustion conditions in the high altitude of Denver) and kitchens with gas stoves (unexpected) have significantly higher average microenvironmental concentrations in Denver than in Washington (Ott *et al.* 1992).

Passengers' CO exposure frequency distributions and factors affecting passenger exposure levels were studied in Koushki *et al.* (1992) in a field experiment in Ridyah, Saudi Arabia. Median exposure level was about 30 ppm CO, and the exposure level remained below 55 ppm about 95 % of the time. The factors that most increased the passenger exposure level were in addition to smoking in order of importance increased traffic volume, decreased average speed, time of day, wind speed and increased interruptions. In a Hong Kong study the CO exposure levels of commuters in busses on the street were 2-3 times higher than at roadside sites (Chan and Wu 1993).

CO & VOC:

Gilli *et al.* (1994) studied the environmental levels and personal exposures on non-smoking university students to CO, benzene, toluene and xylenes in the Piedmont region in Italy, and found that all these aromatics are closely correlated with each other, and that especially xylenes and toluene are also closely correlated with CO. Dor et al (1995) studied the exposures of commuters in Paris to CO and monocyclic aromatic hydrocarbons. Their results were similar to those of Gilli *et al.* (1994), i.e. they found that in automobile transportation, and also e.g. in the Tour St-Jacques ambient air quality monitoring station, the exposure levels of CO, benzene, toluene, ethylbenzene, xylenes and 1,2,4-trimethylbenzene are all closely correlated indicating the same source - motor vehicles. Of the different groups of commuters the automobile drivers and passengers received the highest CO exposures, the subway passengers the lowest and pedestrians and bus passengers found themselves in the middle ground.

Exposure-Response

CO reacts 210 times more strongly with blood haemoglobin than O_2 to form carboxyhaemoglobin, COHb. The high acute toxicity of CO is due to this fact. COHb levels of 1-2 % are harmless and normal for non-smoking urban adults. But only slightly above this level symptoms appear, such as decrease in work time before exhaustion (2.3 - 4.3% COHb), and shortened exercise before pain with angina pectoris patients (2.9 - 4.5 % COHb) (WHO 1987). COHb levels above these are very unlikely to occur in urban ambient outdoor air exposures. But in motor vehicles and indoor environments with indoor sources, such as gas stoves, fireplaces, and internal combustion engines, the CO levels can reach sufficiently high levels to cause acute CO poisoning symptoms and even death. Acute CO poisonings kill annually thousands of people in Europe and much more in the rest of the World. Most of these fatal CO poisonings occur indoors with unvented fireplaces, gas appliances and running engines.

There are only few studies linking increased ambient air CO levels with increased mortality or morbidity. The APHEA study in Athens found a significant association between an increase of ambient 24 h CO level by 10 mg/m^3 and increase of total mortality by 10% (95% CI; 5 - 15%) (Touloumi *et al.* 1996). In a Canadian time-series studies among elderly individuals, the relative risk for hospitalization due to congestive heart failure increased by 6.5% (2.8-10.4%) for a 2 ppm increase in the daily average CO concentrations (Burnett, R, Dales, R *et al.*,1997), confirming an earlier time-series study from sever U.S. Cities (Schwartz, J &Morris, R,1995).

Sampling and analysis

CO can be continuously monitored by lightweight PEMs based on electrochemical cells that oxidize CO into CO_2. Such devices have been used in personal exposure research since mid '70s (Cortese and Spengler 1976, Ackland *et al.* 1985, Jungers *et al.* 1985, Flashbart *et al.* 1987, Ott *et al.* 1988, Wallace *et al.* 1988, Alm *et al.* 1994). From their original bulk, weight, rather small data storage capacity (113 measurements), short operating times (24 h), and zeroing/calibration needs 1 to 4 times each day (Cortese and Spengler 1976, Ackland *et al.* 1985), the personal CO monitors have developed into the size and weight of a Walkman cassette player, data storage capacity of 32,265 values, and operating times of months (Langan 1991). Passive CO sampling tubes have also been developed and tested (Lee *et al.* 1992b and 1992c). They are lightweight, sensitive, selective, cheap and simple, but we have not yet found reports about other studies where these devices have been applied.

1.5.3. Volatile Organic Compounds (VOC)

Definitions

The term VOC is used for a large number of volatile organic compounds with highly varying physical, chemical and toxic properties. The VOC are defined by WHO as having melting points below room temperature and boiling points from 50-100°C to 240-260°C. Other more detailed classifications are in use. The VOC, detectable in a single building, may consist of hundreds of different organic compounds, which makes analysis, risk assessment and guideline setting for these compounds an exceptionally difficult task.

Sources

Traffic fuels:

Probably the most significant and widespread source of VOC exposure is the use of gasoline. Primary gasoline components from evaporation losses during refuelling and from hot engines in the garages, and partly oxidized compounds from tailpipe emissions are both important. The average personal TVOC exposures during refuelling in 6 studies range 50 - 150 mg/m^3. The main components in gasoline vapour are n-butane, isopentane, n-pentane and isobutane. The main components in tailpipe emissions are methane, toluene, ethylene, m, p and o-xylene, n-butane and benzene. The total hydrocarbon levels, (of which only a part comes from gasoline evaporation or traffic emissions) in seven U.S. cities between 6 and 9 o'clock range 200-1400 ppbC. At the component level, i-pentane, n-butane, n-pentane, toluene, m and p-xylene were present at the highest concentrations in all but one city. The mean microenvironmental concentrations measured in automobiles while driving into New York City were toluene (26-56 μg/m^3), m and p-xylene (16-23 μg/m^3), methyl pentane (4-18 μg/m^3), and benzene (9-11 μg/m^3). In addition to the numerous "natural" gasoline compounds, most of which exist at concentrations above and below 1% in the total gasoline, oxygenated compounds, such as methanol, ethanol and MTBE, and many (volatile) organic additives are used to improve the gasoline properties (anti-knock, anti-oxidants, metal deactivators, anti-rust and anti-icing agents, lubricants, detergents and dyes). (Wixtrom and Brown 1992, Weisel *et al.* 1992)

Indoor materials:

In indoor environments harmful VOC are emitted by indoor combustion sources; tobacco smoke, unvented kerosene heaters and sometimes leaking fireplaces (benzene, xylenes, toluene, 2-butanone (MEK)), wood and particleboard (formaldehyde), and a large number of man made building materials, paints, adhesives, gaulking compounds and floor coverings (benzene, xylenes, toluene, styrene, toluene diisocyanate, ethylbenzene, benzyl chloride, 2-butanone, 4-phenylcyclohexene) and a multitude of consumer products for cleaning and maintenance (1,4-dichlorobenzene, trichloroethene, petroleum distillates) (Maroni *et al.* 1995). Typically the indoor air VOC concentrations from fresh paint and carpets, which may initially form an irritating problem, decay to steady state levels within 10..20 weeks from the application (Levin, 1996), and remain low for years before new renovation takes place. The VOC emissions from combustion sources and consumer products may cause more significant long term exposure hazards, because they are used in the same premises repeatedly or continuously.

Activities:

The personal VOC exposures are not only related to certain microenvironments but also to activities, such as painting (xylene, ethylbenzene, decane, undecane and benzene), use of engine cleaner (xylene, ethylbenzene, tetrachloroethylene), household cleaning, or visiting dry cleaners (1,1,1-trichloroethane,

tetrachloroethylene), use of deodorizers (dichlorobenzene), washing dishes or clothes or swimming in a pool (chloroform), auto repair, smoking, pumping gas or driving (benzene, xylene, ethylbenzene), and most importantly many works in many occupations (Wallace *et al.* 1989).

Exposures and Microenvironmental Levels

I am aware of only two studies, where personal VOC exposures of representative population samples have been measured, namely the TEAM studies in New Jersey and California, where the samples were collected more than 10 years ago (Wallace *et al.* 1986, Hartwell *et al.* 1987) and the indoor air and exposure study in Germany (Hoffmann *et al.* 1996). In two new studies, NHEXAS in the U.S and *EXPOLIS* in Europe, VOC exposures will be assessed together with other pollution exposures. There are a few more studies, where the personal VOC exposures of small numbers of people have been monitored in special microenvironments (mostly traffic). All these studies are summarized in Annex I: Table 1.. In order to get a broader idea about the VOC exposures of people one needs to also look at studies where VOC have been analysed in typical microenvironments, such as homes, office buildings, autos, streets, and urban outdoor air.

Table 6. in Annex I presents the health risks and some reference values for 59 different VOC (Health effects will be discussed in a later chapter) and Table 7. presents the available personal and microenvironmental concentration data from 28 recent microenvironmental and personal exposure studies for the listed VOC.

A close evaluation of the data on Tables 6. and 7. reveals that the list contains 59 VOC that have some reference value based on health, odour or irritation. 12 of these 59 VOC were not analyzed in any of these 28 studies. Out of these 12 vinyl chloride is a known carcinogen, methylisocyanate is a strong airway hypersensitívity initiator, a skin contact allergen and a strong irritant, methylmetacrylate is a skin contact allergen, and acetic acid an irritant. 11 of these 59 VOC have been measured in only one study. The whole VOC list contains 29 of the 172 organic U.S.Clean Air Act Amendments of 1990 (CAAA 1990) listed Hazardous Air Pollutants (HAPs), 2 of which (vinyl chloride and methylisocyanate) have not been analyzed in any of these studies.

Concluding from the evaluation of the 28 recent VOC studies: The sets of VOC that have been selected for analysis in different studies are inconsistent, and it is not clear from most reports what the selections are based on. Often the selection seems to be more related to available equipment, convenience and experience than consideration for the relevant compounds either for the complex mixture in the air or for the health effects of concern.

In a review of 68 VOC studies Brown *et al.* (1994) conclude that:

a) The mean concentration of each VOC in established buildings is generally below 50 $\mu g/m^3$, with most below 5 $\mu g/m^3$, while TVOC concentrations are substantially higher (e.g. 1100 $\mu g/m^3$ in dwellings) reflecting the large number of compounds present;

b) The mean VOC and TVOC concentrations in dwellings are generally greater than those in established public buildings, for unknown reasons;

c) VOC concentrations in new buildings are much greater than those in established buildings, often by an order of magnitude or more, and appear to arise from construction materials and building contents, the VOC emission characteristics of which can be measured for source control; and

d) VOC and TVOC concentrations in complaint buildings have been measured to a limited extent and may or may not be greater than those in established buildings.

At least two studies have addressed systematically the TVOC levels in building stock. In the Swedish ELIB study TVOC was measured together with a number of other IAQ and energy parameters in a carefully selected representative sample of the Swedish housing stock. They found an essentially log-normal distribution of indoor air TVOC levels with a median value between 300 and 400 $\mu g/m^3$, single-family houses slightly higher than multi-family buildings (Norlén and Andersson 1993). In the European AUDIT project of 56 office buildings in 9 countries, differences in the TVOC levels between countries were striking. In Germany, Finland, the Netherlands and Denmark the TVOC levels varied 100 - 400 $\mu g/m^3$, while in the U.K, Greece and France the range was 200 - 1000 $\mu g/m^3$, in Switzerland and Norway the levels were generally low but peak values reached 1800 $\mu g/m^3$ (Levin, personal communication 1996).

Exposure-Response

The health effects of different VOC are quantitatively and qualitatively quite different. Table 6. in Annex I lists the VOC compounds that have been assigned reference or guideline values such as odour (Devos 1990, USEPA database) and irritation (WHO 1987, AT 1992, Schaper 1993) threshold levels, air quality guideline values (WHO 1987, ASHRAE 62 1989), or are listed as Hazardous Air Pollutants in the U.S.Clean Air Act Amendments of 1990. Environmental measurements have been published for 103 of the 172 organic HAPs. All the 172 organic HAPs are not listed in Table 6., but those 29 that are in the list, have been marked in the last column. Some VOC are known or suspected human carcinogens, see column 4 in Table 6. (IARC 1987), airway hypersensitivity initiators, see columns 2 and 3 in Table 6. (Bakke *et al.* 1993, NKB 1994), irritants and/or odorous, see columns 5-11 in Table 6. (Devos, 1993, Schaper, 1993, AT 1992, WHO 1987).

The known human carcinogenic VOC (IARC I) are benzene and vinyl chloride, and the suspected carcinogens (IARC II A, B) are formaldehyde, 1,3-butadiene, styrene, tetrachloroethane, chloroform, hexachlorobenzene, and chlorophenols. A recent U.S. study suggests that 1,3-butadiene makes the greatest contribution to overall VOC related cancer risk, namely 41 %, followed by 18 % for benzene and 15 % for formaldehyde. Yet, these are not likely to be the most significant cancer risks in air environments. Most of the organic carcinogens that occur in the indoor environments are particle phase PAH:s from combustion sources, such as tobacco smoke, fireplaces and diesel exhaust gases from outdoors, and the dominant indoor air carcinogen is most probably the radioactive noble gas, radon (^{222}Rn) emanating from some building materials and the soil beneath the buildings.

No VOC occurring in indoor air are known allergens in the sense that they would cause Ig-E transmitted immunological defense mechanisms in the organism. However, some indoor air VOC satisfy the definition for specific hypersensitivity initiators, which alter enzyme function or metabolism in the organism and can, in sensitized individuals, cause strong reactive symptoms at low doses. Once an individual's airways have been sensitized by allergy or hypersensitivity they are typically also quite sensitive to non-specific hyper-responsiveness caused by quite many irritating VOC, particles, smokes and their mixtures (Bakke *et al.* 1993, NKB 1994).

Some VOC exhibit neurotoxic effects (xylenes, toluene, styrene, trichloroethane, ethyl benzene, dichloromethane, 1,4 dichlorobenzene, benzyl chloride, 2-butanone, 4-phenylcyclohexene and many petroleum distillates). Occupational toxicology and epidemiology, and animal tests have also identified other health relevant properties for some VOC. However, all these effects are only known to occur at concentrations which must be quite rare in non-industrial indoor environments.

In the high end of the VOC exposures in buildings the general human response to VOC in indoor air has been classified to A) acutely perceived deterioration of the environment, B) acute or subacute

inflammation-like reactions in skin or mucous membrane, and C) subacute and weak stress-like reactions.

It has been suggested that there may be an association between rather low TVOC levels with a complex set of unspecific symptoms, called the Sick Building Syndrome (SBS). However, in evaluating this connection Mendell found in his review (1993) of 33 studies on the environmental factors related to the SBS (Sick Building Syndrome) only sparse or inconsistent association between the observed VOC levels and work related symptoms. One possible explanation for this lack of association may be the lack of consistency in the VOC measured in different studies, discussed in chapter **Microenvironmental levels and personal exposures**. One of the most interesting recent findings is that the irritating (nasal pungency) effects of many VOC in complex mixtures appear to be proportional to their odour effects and additive and can be modelled in some cases (Cometto-Mu iz and Cain 1995). When one compares the no observed effect levels (NOEL) for individual VOC to the levels found in (complaint) buildings, it becomes obvious that any acute health effects or symptoms that the existing VOC levels (Table 7. and Brown *et al.* 1994) in buildings might exhibit must be some types of combined effects.

Perceived air quality:

People perceive the VOC by their odorous and common chemical sense (chemesthetic modality). Several methods have been developed to evaluate the combined perceived effects of mixtures of VOC and other air quality deteriorating compounds. The most studied and developed is probably the Decipol method, which uses trained panels, calibrated by known concentrations of acetone (decipol scale) to rate their first impact of perceived air quality e.g. in a room. This method has left few people cold, it has raised both enthusiasm and strong criticism (Bluysen and Elkhuizen 1996, Aizlewood *et al.* 1996A and B, Fanger 1996), depending on the variation of viewpoints from practical relevance to scientific accuracy. On the cool side a typical conclusion from the discussion has been that the decipol rating does not replace existing IAQ monitoring techniques or measures, but may add a practical dimension for systematical evaluation of IAQ - which can be compared as a method and purpose to tasting drinking water after all the chemical tests have been passed. Obviously, if the water tastes awful, its quality is unacceptable regardless of other test results, but will a pleasant fresh taste give it high quality in the absence of chemical tests?

TVOC approach:

In spite of the studies of Cain *et al.* (Cometto-Muñiz and Cain, 1995) and the radical approaches by Fanger *et al.* (1988, Bluysen *et al.* 1991) science and practice are still far from predicting the combined health and sensory effects of complex VOC mixtures. Facing great uncertainties decisions still need to be made daily in selecting materials and ventilation rates to new buildings and judging problems in existing ones.

Two practical approaches for IAQ guidelines for total VOC (TVOC) (excluding formaldehyde and carcinogenic VOC) have been proposed, one for total VOC measurement (TVOC) (Molhave 1990), the other based on gas chromatographic separation and quantification (Seifert 1990).

The former (Molhave 1990) approach is generalized from the toxicological responses published in indoor air pollution literature. The following exposure range classification relative to the TVOC-level as measured by flame ionization detector calibrated against toluene is suggested: Comfort range (< 200 $\mu g/m^3$), multifactorial exposure range (200-3 000 $\mu g/m^3$), discomfort range (3 000-25 000 $\mu g/m^3$), and toxic range (> 25 000 $\mu g/m^3$).

In the latter (Seifert 1990) approach, the analyzed organic compounds are ranked according to their concentrations and divided into the following classes (class target guideline for ten first in each class in

parenthesis): alkanes (100 μg/m^3), aromatics (50 μg/m^3) terpenes (30 μg/m^3), halocarbons (30 μg/m^3), esters (20 μg/m^3), carbonyls (excl. formaldehyde) (20 μg/m^3), and "other" (50 μg/m^3). The classes are then summed up for the TVOC-value. The proposed target guideline value for the TVOC is (300 μg/m^3), and no individual compound should exceed 50% of its class target or 10% of the TVOC target guideline value. These target guideline values are not based on toxicological considerations, but on the existing levels and on professional judgement about the achievable levels.

Although the two approaches are fundamentally different, they almost agree in the practical outcome. The first suggests a comfort range of < 200 μg/m^3, the latter proposes a target guideline value of 300 μg/m^3 for the TVOC.
For those interested in further development of the TVOC concept, Working Group 13 of the European Concerted Action "Indoor Air Quality and its Impact on Man" has prepared a new guide for analysis and application of the TVOC (1997).

Sampling and Analysis

VOCs can be sampled by passive personal or microenvironmental tubes or badges containing solid absorbents, mostly Tenax (like in Fellin and Otson 1993, Otson *et al.* 1994, Otson and Fellin 1993, Crump and Madany 1993, Stridh *et al.* 1993, Cao and Hewitt 1993, Matsumura *et al.* 1993), but also evacuated canisters (Chan *et al.* 1991, Highsmith *et al.* 1993) and charcoal or multisorbent media (Faust *et al.* 1993, Bayer *et al.* 1993, Cottica *et al.* 1993) have been used. The diffusive sampling flows and sampling efficiencies of the passive sampler vary for different VOCs (Cao and Hewitt 1993). Consequently the VOC concentration ratios in the absorbent are not the same as the VOC concentration ratios in the sampled air. Yet, with sufficient additional information the concentrations in air can be recovered from the sample analysis data. If the sample flow is pumped through an absorbent containing sampling tube (like in Hartwell *et al.* 1987, Chan *et al.* 1991, Chan *et al.* 1993a, Chan *et al.* 1993b, Crump and Madany 1993, Saarela and Mattinen 1993, Saarela and Mattinen 1993, Saarela *et al.* 1993, Singhvi *et al.* 1993, Nieslochowski 1993, Rothweiler *et al.* 1993, LoSurdo *et al.* 1993, Op't Veld 1993) this problem disappears, but the sampler becomes heavier, more expensive and the sampling time is limited by life of the battery.

The total VOC (TVOC) is often calculated as a sum of all measured individual VOCs. Considering how different the sets of VOCs are that have usually been selected for personal exposure or microenvironmental concentration studies, such summed TVOC-levels are not directly comparable to each other. Additionally, TVOC concentrations have been measured by a range of definitions of unknown relationship to each other, limiting interpretation of exposure assessment by this measure (Brown *et al.* 1994).

Continuous VOC monitoring:

TVOC can be measured directly and continuously (Ekberg 1993, Lee 1993d by photoacoustic spectrometry (PAS), non-dispersive infrared spectrometry (NDIR), Fourier transform infrared spectrometry (FTIR), or flame ionization detection (FID). While the VOCs that have been sampled on solid absorbents and analyzed by GC with different detectors necessarily represent levels averaged over the whole sampling time, direct methods have the advantage of being capable for continuous monitoring. Because the molecular weights of different VOCs vary considerably, the direct methods need to be calibrated against and expressed as some defined VOC compound (or mixture).

In a study of Ekberg (1993) continuous TVOC monitoring by PAS enabled direct real time comparison of indoor air TVOC levels with outdoor air TVOC and CO levels and time patterns. The comparison of the time patterns (which is not possible with time integrating measurement methods) led to the conclusion that the traffic generated outdoor VOCs were responsible for 60-90% of the indoor TVOC

level. Lee *et al.* (1993d) compared the applicabilities of the FID, NDIR and FTIR techniques for continuous indoor air VOC monitoring. Lee concluded that FID and NDIR can only be used to follow increases and decay of (T)VOC levels with time, and therefore can be used to help identify sources and sinks or dilution mechanisms. NDIR was found to be less useful than FID. The FTIR proved to be very useful and effective for VOC monitoring, and could be used to identify specific VOCs and their sources. Yet, even FTIR was not particularly useful for monitoring and identifying a mixture of many VOCs.

Continuously recording VOC monitors are becoming available but they are not yet suitable for personal monitoring.

1.5.4. Nitrogen Dioxide

Sources

NO_2 is produced as a primary air pollutant in high temperature combustion with plenty of excess air, like in gas stove flames and diesel engines. Most of the NO_2 in ambient air is secondary air pollutant produced in the atmosphere by the reaction of NO (from traffic exhaust, heat and power production) with O_3. In urban air this reaction is usually limited by the availability of O_3, which is slowly mixed into the NO-polluted boundary layer from the higher and more stable layers of the troposphere. While the level of NO in the urban air varies greatly in time and space, the level of NO_2 is more evenly spread over large areas. In the absence of indoor NO_2 sources, the indoor level of the reactive NO_2 is usually only a fraction of the outdoor level. The dominant indoor sources of NO_2 are gas stoves and unvented gas fired water heaters. Such indoor sources may significantly contribute to the individual NO_2 exposures (e.g. Avaliani *et al.* 1993, Dörre and Knauer 1993, Song *et al.* 1993, Xue *et al.* 1993, Özkaynak *et al.* 1993).

Exposure

Exposures of children:

In a study on the personal exposures of preschool children in Helsinki to air pollutants, the one week average NO_2 exposures of 246 children were measured with personal Palmes tubes, which the children carried in their clothes (Alm *et al.* 1995, 1998). Researchers found that the effect of gas burning stoves (mode 25-30 $\mu g/m^3$) vs electric stoves (mode 20-30 $\mu g/m^3$) on the average personal NO_2 exposures was smaller than the effect of downtown (mode 20-30 $\mu g/m^3$) vs suburban (mode 15-20 $\mu g/m^3$) residence. NO_2 exposures of still younger children, 2-3 years of age, were assessed indirectly in a German study using activity recording and a microenvironmental model. The children were all living in Berlin and went to child care facilities during their parents= working hours. The NO_2 sources were outdoor air and indoor gas appliances. The individual total modelled 24 h NO_2 exposures varied 1:3. Also the contributions of all microenvironments to the modelled total exposure varied greatly. In general exposures inside the homes and the child care facilities gave an overwhelming contribution to the total exposure, although the NO2 concentrations in the traffic and outdoors were higher than the indoor concentrations - when no unvented gas appliances were used. (Dörre and Knauer 1993)

Among 623 Swiss children, aged 0-5 years, NO2 levels have been assessed with passive samplers for a 6-week period. In all four regions, average NO2 concentrations measured outdoor at home where 1.6 to 2.3 times higher then those indoors (Braun-Fahrländer *et al.*,1992).

The personal NO_2 exposures of newborn infants were determined in Oslo using personal 14 day Palmes tubes plus stationary Palmes tubes in each infant's bedroom, living room, kitchen, and outdoors at home

address. Time activity recording shows that the newborn infants spend as an average less than 50% of their time outside of their own bedrooms and 83% of their time in their own homes. Not surprisingly their personal exposure levels are almost identical to the home indoor air levels and well below the outdoor values. There are no gas stoves in Oslo. (Oie *et al.*1993)

Exposures of adults:

In the studies on adult exposures NO_2 the three major contributors are gas stoves, traffic, and outdoor air. In different studies the contribution of the gas stoves has varied from quite considerable (18-36 $\mu g/m^3$ above outdoor levels, with and without pilot lights), like in the study on 350 individuals in Portage WI (Quackenboss *et al.* 1986), to significant (without pilot lights10 $\mu g/m^3$ above homes with electric stoves) like in the Boston study on 313 individuals (Ryan *et al.* 1989), to moderate (without pilot lights 5 $\mu g/m^3$ above homes with electric stoves), like in the Los Angeles Study on 682 individuals (Spengler *et al.* 1994). When the homes with gas stoves having pilot lights were separated from those without pilot lights, the contribution of pilot lights to the indoor NO_2 level was significant (Ryan *et al.* 1989, Spengler *et al.* 1994).

Exposure to NO_2 in road traffic was found to be 2 - 4 times higher than ambient outdoor air in Nottingham, U.K. (Catward and Colls 1990), and about 1.5 times higher in Hong Kong (Chan and Wu, 1993).

Exposure-Response

The new draft WHO Air Quality Guideline values for NO_2 are 200 $\mu g/m^3$ (1 h) and 40-50 $\mu g/m^3$ (24 h) (WHO 1995). However, recent research has shown that NO_2 levels, much below these guideline values, may have significant effects on human health. In a large American cross sectional study covering tens of cities, a significant association was found between the annual mean NO_2 level and lung function of 6-24 year olds. The reduction of the lung function was most significant for cities, where the annual average NO_2 level exceeded 75 $\mu g/m^3$ (Schwartz 1989).
In Helsinki, variation of the 24 h ambient air NO2 levels from 4 to 170 $\mu g/m^3$ was associated with significant increases in hospitalization of adult asthmatics (Pönkä 1990 and 1991). In 5 German cities, variation of the 24 h NO_2 level from 10 to 70 $\mu g/m^3$ was associated with a 28 % increase in the prevalence of laryngitis in children (Schwartz *et al.* 1991). In Holland, variation of the 24 h NO_2 level from 2 - 70 $\mu g/m^3$ was weakly associated with reduction of the lung function of children (Brunekreef *et al.* 1989, Hoek 1992). In a large Swedish study, the variation of the mean wintertime NO_2 level (10-32 $\mu g/m^3$) was found to be significantly associated with respiratory symptoms (Forsberg *et al.* 1991).

On the other hand, a yet unpublished European epidemiological study (SAVIAH), based on superior personal ambient NO_2 modelling in four European cities shows no effects of ambient NO_2 level on respiratory symptoms in children. In the European APHEA study, a time series study on the associations of urban air pollution levels and mortality and morbidity, NO_2 levels (29-86 $\mu g/m^3$) had a non-significant positive effect on respiratory emergency hospital admissions in Rotterdam (Schouten *et al.* 1996), NO_2 levels (4-324 $\mu g/m^3$(!)) had no effect on mortality in Lyon (Zmirou *et al.* 1996) or Köln (NO_2 levels 24-82 $\mu g/m^3$) (Spix and Wichmann 1996), asthma admissions correlated with NO_2 levels (22-108 $\mu g/m^3$) in Paris (Dab *et al.* 1996), but not with NO_2 levels (33-41$\mu g/m^3$) in Helsinki (Pönkä and Virtanen 1996).

In the large population based Swiss Study on Air Pollution and Lung Diseases in Adults SAPALDIA, adjusted lung function (FVC and FEV1) where significantly correlated with long-term average NO2 levels across the eight study areas (Ackermann-Liebrich *et al.*,1997). This has also been confirmed with a within-area analyses, using personal and neighbourhood levels of NO2 exposure rather then just data from one fixed site monitor (Schindler *et al.*, 1998). The Swiss study among children, SCARPOL,

observed a significant association of repeated cough, pneumonia and bronchitis with the long-term average level of air pollution (Braun-Fahrländer *et al.*, 1997).

It is important to note that epidemiological studies have inherent difficulties to partition the independent effects of single pollutants in the urban air pollution mixture. This is particularly true for pollutants which correlate highly due to common sources. Therefore, NO_2 may not be the causative agent *per se*, but an excellent indicator of traffic and photochemistry related air pollution.

Personal Sampling and Analysis

NO_2 is the most measured individual pollutant compound in personal exposure studies. The development of the cheap, sensitive and simple Palmes tube in 1976 (Ref in Quackenboss, 1982) opened the way to affordable personal exposure and microenvironmental monitoring at 7-14 day averaging times. Since the Palmes tube, other passive NO_2 badges have become available that shorten the averaging times to 24 h or less (Yanagisawa *et al.* 1982 (see Lee *et al.* 1992a)) have been tested for various sampling purposes (Lee *et al.* 1992a, 1993a, 1993b) and are more specific to NO_2 alone than the Palmes tube (Spicer *et al.* 1993). Continuously recording portable or personal NO_2 monitors based on electrochemical cells are also presently available. However, due to difficulties in data interpretation, possibly because of some significant but unknown interferences, little or no data that has been produced by such devices has been published so far (possibly in Schauer & Dörre 1993).

2. OVERALL DESIGN OF *EXPOLIS*

2.1. Scope and Objectives of Expolis

The EXPOLIS (Air Pollution Exposure Distributions within Adult Urban Populations in Europe) study focuses on working age urban populations in Europe, exposed to air pollutants in their homes, workplaces and other common urban microenvironments (streets, shopping, etc), and commuting between them. Significant occupational exposures originating from specific work processes to the pollutants of interest are excluded, because the affected individuals in this survey are likely to be too few for reliable statistics. Occupational exposure statistics from other sources should be used if such occupational exposures are wanted in the simulation of the population exposures. The included occupational exposures are related to work done in offices, educational facilities, health care and nursing facilities, supermarkets, outdoors and in transportation.

In this first stage of database build-up the measured pollutants are $PM_{2.5}$, CO and VOC. This microenvironmental concentration/exposure frequency distribution database can later be expanded to include NO_2, aldehydes, radon, other ionizing radiation, different population subgroups, etc. The urban areas selected for the *EXPOLIS* study are Athens, Basel, Grenoble, Helsinki, Milan and Prague, to represent different European regions, city sizes and air pollution situations.

The general objectives of this study are:

- To measure the exposures of adult urban populations to major air pollutants, and some key parameters that affect these exposures.

- To improve environmental health risk management by developing a technique for assessing and predicting the air pollution exposure consequences of alternative urban development policies.

The specific goals of this study are to determine:

- Frequency distributions and other basic statistics of the exposures of European adult urban populations to air pollutants; volatile organic compounds (VOC), carbon monoxide (CO) and respirable particulate matter (PM2.5),

- Distributions of the timing and amounts of time spent by adult urban populations in different microenvironments,

- Roles of geographic, housing and commuting related, behavioural and socioeconomic risk factors in the air pollution exposures of adult urban populations in Europe,

- Roles of different air pollution sources in the air pollution exposures of adult urban populations in Europe, and to prepare

- European databases for the simulation of air pollution exposures of the urban populations at large, selected subpopulations, and populations in alternative future exposure scenarios.

The original idea of EXPOLIS was to sample each individual 3 times, but with the budget fixed the advantages of repeated measurements had to be balanced against the disadvantages of very small sample sizes and consequently the representativeness of the population samples was considered more important than the ability to compare within and between individual differences.

2.2. Study Sites

Exposures and microenvironmental concentrations of selected major air pollutants, $PM_{2.5}$, CO and 30 VOCs, were measured in six European cities: Athens, Basel, Grenoble, Helsinki, Milan and Prague, see Figure 2.2.-1. These cities were selected to represent different European regions, climates and populations. Selection was also dictated by the presence of a research facility capable and willing to carry out this study protocol.

Athens is the capital and largest city of Greece. It lies on a small plain that extends southward to the Aegean Sea. The city Centre is 11 km from the coast. Greater Athens has a population of over 3 million - 1/3 of the population of Greece. Athens has a typical Mediterranean climate, with hot, dry summers (> 25°C) and mild winters (10°C). Average annual rainfall is 400 mm.

Basel is located in northern Switzerland. The population of the city with suburbs is nearly 400,000. Located on the Rhine River, Basel is a major Centre of chemical and pharmaceutical industry and commercial port. Rainfall averages 1,000 mm per year, and moderate number of days with fog in the in winter. The average temperature ranges from 0.6°C in January to 18°C in July.

Grenoble is the Capital of the French Alps about 100 km South-East of Lyon on the Isere River. The Metropolitan area population is over 400,000. Hydroelectric power from Alpine rivers provides much of the energy for the production of electrical machinery, electrometallurgy, cement, chemicals, and plastics. The climate is characterized by warm, dry summers (20°C) and relatively mild winters (0°C), with an average annual rainfall of 1000 mm.

Helsinki is the capital and largest city of Finland. It is located on the southern coast of the country on the Gulf of Finland. The population of Helsinki metropolitan area is about 1 million. Helsinki is Finland's chief port and handles more than half of all its foreign trade. Engineering electronics and shipbuilding industries and food and timber processing are important. Climate exhibits both maritime and continental influences. Surrounding seas cool the climate in spring but warm it in fall. Rainfall averages 700 mm per year. The sea is frozen and the ground covered with snow for several months each winter. Mean temperature in January is -6°C, but the summer months are mild (17°C).

Milan is the capital of Lombardy. The population of Milan metropolitan area is nearly 4 million. Milan is located in the basin of the Po River about 480 km northwest of Rome. Most industrial development has taken place in Milan's suburbs, far from the central city. Milan is Italy's chief commercial, financial, and industrial Centre manufacturing steel, textiles (particularly silk), clothing, machine tools, aircraft, automobiles, railroad equipment, agricultural machinery, chemicals, printed materials, pharmaceuticals, furniture, and foodstuffs. Milan has a continental climate. Seasonal temperatures average 24°C for July and 5°C for January.

Prague is the capital and largest city of the Czech Republic. Its population is 1,200,000. The city is situated along both banks of the Vltava River. It is an industrial city, producing goods ranging from machinery, rolling stock, and chemicals to textiles, furniture, foodstuffs, and beer. Winters are generally

cold, with many days of subfreezing weather (January average -1°C). Summers are moderately warm with average July temperature about 19°C.

In each city, a population sample of 25-55 (Grenoble 20-60) year old persons was formed and subsamples for exposure measurements and questionnaire applications were drawn. Population sample sizes are summarized in Figure 2.2.-2. and Table 2.2.-1.

Identical time activity and background questionnaires were used for *exposure sample* and *diary sample*. Personal exposures as well as the most important microenvironmental concentrations were measured for the exposure sample. The microenvironments investigated were home indoors, home outdoors and main work place.

In each city, also a selected group of public microenvironments were measured. These microenvironments include shops, restaurants, indoor sports facilities and public transport.

2.3. Air Pollutants

In each Centre, the personal exposures and personal microenvironmental concentrations were measured for $PM_{2.5}$, CO and 30 VOCs (see Table 2.3.-1.). The major air pollutants common to all cities were selected based on their health effects and their environmental concerns as follows:

- CO to represent exposure to traffic exhausts and indoor combustion sources,
- VOCs (see Table 2.3.-1.) because of health and welfare concerns both indoors and outdoors (carcinogenic, odorous and irritating compounds, precursors for tropospheric O_3), because many VOCs are useful source markers, and because the presently available data are of very variable quality, and
- $PM_{2.5}$ because inhalable particles are presently the air pollutants of greatest health concern and interest, and because no $PM_{2.5}$ exposure studies on representative population samples have been reported so far.

In addition 48 h NO_2 samplers were collected from Basel, Helsinki and Prague. In Helsinki also carbonyl compounds and air exchange rate, and in Milan aldehydes were measured. Table 2.3.-2. summarizes the air pollutants, microenvironments and measurement techniques of *EXPOLIS*.

2.4. Microenvironments and Activities

A microenvironment (ME) is a location where, for the purpose of the study, the air pollutant concentrations at any given time can be considered homogenous. For population exposure distribution simulations, all individual microenvironments that fall into the same category are grouped and processed as one microenvironment, and the concentrations measured or modelled for this microenvironment are presented in the form of a frequency distribution. In air pollution exposure modelling and simulation, concentration information from the microenvironments contributing significantly to the population exposure is needed. The microenvironments selected for the EXPOLIS time activity diaries were *home*

indoors, *home outdoors*, *workplace indoors*, *other outdoor* and *other indoor*, and *traffic* (with subcategories), see Figure 2.4.-1.

Work environments differ more than home environments from the viewpoint of exposure to air pollution. Public services, shops, offices, industrial work, transportation all have different characteristics. Heavy occupational exposures are excluded from the analysis, because they are too uncommon to be adequately represented in our population samples.

The microenvironments/activities, about which information was separately collected, were transportation (with subcategories), supermarkets, indoor sports facilities, public buildings and restaurants in order to assess average exposure levels of $PM_{2.5}$, VOCs or CO in these locations. Microenvironmental concentrations in traffic - inside automobiles, busses, trams, trains, metros and while walking or biking - were measured separately during the most active traffic hours. Microenvironmental levels in supermarkets and restaurants were measured during their active opening hours. Exposures related to specific activities were measured by the field team members.

2.5. Target Populations

The target populations of this study are the adult, urban populations of Europe. *EXPOLIS* focuses on 25-55 (20-60) year old individuals, because their exposures are most affected by urban traffic planning, zoning and occupational conditions. Individuals participating in the study should live and work in the target area and they must not travel too much. WHO (1991) estimates that a probability sample of a minimum of 50 subjects are needed for the sample to represent any target population. Larger samples are needed if the target population is divided into subpopulations for quantitative estimation of how the exposures relate to, e.g., home location, indoor sources, commuting, work, and socioeconomic parameters.

Too small subsamples produce poor estimates about exposure frequency distributions in the respective subpopulations. On the other hand, ensuring that all interesting subpopulations would have at least 50 representatives in our probability samples in each of the six *EXPOLIS* cities would result in a prohibitively expensive study.

2.6. Measurement scheme

The personal exposure and microenvironmental concentration data were collected from the *Exposure* subjects during one year from summer of 1996 to winter of 1997-98. Each subject carried a personal exposure monitoring case, and her/his home, inside and outside, and workplace were equipped with microenvironmental measuring equipment for a period of 48 h. If the subject did not work at all or worked at home, the work measurement took place at subject=s home for the normal working hours. The workplace concentrations were measured for the normal working hours at the actual workplace of the subject, or if the subject moved from place to place during work, at a typical workplace. The home inside and outside concentrations were monitored from the time when the subject would normally return from work to the time when she/he would normally leave home for work. Outside concentrations were monitored, if there was a safe balcony/yard or similar outside location next to the apartment of the study subject.

The measurements were made during the work weeks, mostly from Monday morning to Wednesday morning, and Wednesday evening to Friday evening. The *Diary* subject's data collection covers the same periods.

Weekend exposures were considered either (i) to be simpler than workday exposures, and (ii) to occur often outside of the urban area of interest, or (iii) to be too uncommon (e.g. moth spraying, painting, motorbike maintenance), which on the one hand would require much larger population samples for representative coverage and on the other hand are outside the main scope of urban environmental management.

The common weekend exposures can be simulated using the database, the less common ones would require separate focussed sampling programs.

2.7. Personal and Microenvironmental Measurements

The purpose of the following description of the sampling and analysis procedures is just to shortly list the equipment and name the methods. Detailed descriptions of the $PM_{2.5}$ and VOC sampling and sample analysis techniques, together with VOC methods intercalibration data, QA methods and QC data will be published separately.

The personal exposure monitoring equipment (PEM), (sampling pump, 2.5 μm cyclone, 37 mm holders with filters, VOC sampling tube, CO monitor, and a battery pack) was packed into a 5.2 kg (total) aluminum briefcase carried by each subject for 48 h. The modified Buck IH (A.P.Buck Inc., Orlando FL) pump is silent, lightweight and after modification capable of sampling 48 h with a single set of batteries and therefore suitable for personal measurements. It was adjusted to draw air at 4 L/min using a simple volumetric flow control. Small $PM_{2.5}$ GK2.05 cyclones for personal $PM_{2.5}$ sampling at 4 L/min were designed and constructed for the *EXPOLIS* study (BGI Inc., Waltham, MA). With this design the filters are handled from pre- to post-weighing in standard 37 mm plastic filter holders which minimizes the risk of filter contamination and damage in the field. In the laboratory the flow rate was adjusted to 4 L/min with a bubble flowmetre (Mini BUCK Calibrator M-30) before and controlled after the sampling period. Two filter holders with 2 μm pore Gelman Teflo (Gelman Sciences, Ann Arbor, MI) filters were provided for each subject: one 'day filter' for two sampling periods beginning at leaving home for work and ending at return home from work, and one 'night filter' for the remaining times. The subjects changed the PEM filter holders according to personal instructions.

VOCs were sampled into a Perkin Elmer Tenax-TA tube (VOC-tube) by vacuum of the same pump that sampled the $PM_{2.5}$. The target sample size was 2 to 3 L, the VOC-tube flow rate was restricted to about 0.5 - 1.0 mL/min, and VOC diffusion to the tube before and after timed sampling was prevented by drawing the sample air into and from the VOC-tube through 200 mm long stainless steel capillary tubes. VOC-tube flow rate was measured before and after each sampling by a bubble flowmetre (Mini BUCK Calibrator M-1). In Basel the VOCs were collected using Carbotrap tubes instead of Tenax-TA. The target sample sizes and flow rates for Carbotrap sampling were about 10 times higher than with Tenax-TA.

The CO-PEM used was the CO Enhanced Measurer T15 (Langan Products Inc., San Francisco CA) based on diffusion air flow to a CO specific electrochemical detector. The unit records the CO concentration (0.1 - 12.8 ppm, and 1 - 128 ppm ranges) as well as internal and external temperature in short, user-selectable intervals. 1 minute interval was used in *EXPOLIS* measurements. The measured values together with date/time were internally stored in memory for later downloading to a computer.

Workplace and home indoor and outdoor microenvironmental monitors (MEM) (sampling pump, 2.5 μm impactor, 47 mm filter holder with filters and a VOC tube packed into a portable sound absorbing container) were programmed to run inside and outside of the home for the expected non-working hours and in the workplace for the expected working hours of each subject. The MEM sampler contained a WINS PM2.5 (EPA Well Impactor Ninety Six) impactor (BGI), a 47 mm filter holder (BGI) with a Gelman Teflo filter and a PQ100 pump (BGI). The WINS PM2.5 is a single jet well impactor designed to remove particles with a 50% cut size at 2.5 μm at 16.7 L/min. A Graseby-Andersen PM_{10} inlet (Sierra-Andersen, Inc.) preceding the WINS PM2.5 impactor was used in outdoor measurements during bad weather to avoid wind and rain effects. The PQ100 pump is weatherproof, equipped with a microprocessor-controlled timing and mass flow adjustment system, and capable of operating up to 36 h on an internal lead-acid battery. The pump is designed to pull in a sample of air at a constant flow rate of 1.0 - 25 L/min (mass flow rate accuracy ±5 %). The flow rate was measured/adjusted before each sampling and controlled after sampling with a bubble flow metre (Buck M-30).

The VOC-tube arrangement was identical to the PEM case, except that the flow rate was adjusted to about 2 mL/min with Tenax TA sampling and 20 mL/min with Carbotrap sampling.

Sample analysis: For analysis the VOCs were thermally desorbed from the tubes and subsequently analysed at VTT, Chemical Technology, Finland, by GC separation and simultaneous detection by MSD and FID. The VOC samples collected on Carbotrap, were analysed by Carbotech, SA in Switzerland using GC/FID technique. The $PM_{2.5}$ sample filters were weighed before and after sampling in each Centre using a microbalance, and archived in a refrigerator for later elemental/chemical analyses. These analytical procedures together with QA/QC data will be described in detail in later articles.

The NO_2 samples were collected using Palmes passive tubes (Palmes et al. 1976). The tubes were prepared and analysed spectrophotometrically at the Swiss federal Institute of Technology in Z rich. This technique has been used for personal sampling in numerous studies (e.g. Alm et al. 1998) and proven reliable.

Pilot phase

The selected measuring equipment were tested in Milan and Kuopio prior to the pilot. Prior to the survey all equipment, techniques, training, instructions, questionnaires, standard operating procedures (SOPs) and general information materials were tested with volunteer subjects. The pilot samples were analysed and the pilot experiences collected from all EXPOLIS Centres. This material was discussed and assessed in a common workshop, and the SOPs, questionnaires and TMADs were edited according to the pilot experiences. The database structure and data entry (questionnaires and TMAD) and downloading tools (pumps, CO monitors and microbalances) were developed and tested during and after the pilot phase.

13 subjects were included in the Pilot sample in Helsinki, 3 to 5 subjects in the other centres.

2.8. Team Organisation

A complicated multicentre (and multilanguage) protocol like *EXPOLIS*, where multiple compounds are monitored in multiple microenvironments, needs a great deal of practical everyday problem-

solving and other communication to ensure on the one hand a common practice and comparable study results, and on the other hand minimum data losses. The junior researchers were trained at the different phases of the study together in *EXPOLIS*-Workshops in Prague (April 21 - 24, 1996), Helsinki (September 9 - 13, 1996), Grenoble (March 23 - 26, 1997), and Bilthoven (February 5 - 8, 1998). These opportunities were also used for equipment intercalibrations. In each Centre one researcher was assigned to one or more of the following contact groups: *Equipment*, *Database*, *Questionnaires*, *Time-Activity-Diary* and VOCs. QA/QC and *Privacy Protection* responsibilities lay within the principal investigators. For example the Database Contact Group members collected all database-related problems, ideas and experiences in each Centre, and communicated them to other Centres for distribution there. Communication occurred mostly via E-mail and faxes, but each junior researcher was also assigned a GSM telephone (Nokia 2110 or 1610) with the GSM numbers of all other *EXPOLIS* junior researchers and principal investigators programmed to ensure fast access when and where problems/questions were encountered in the field or laboratory.

2.9. FIELD SURVEY

A randomly drawn "base sample" population first received an information letter about *EXPOLIS* and a Short Screening Questionnaire, which they were asked to complete and send back to the local *EXPOLIS* Centre, including the response card indicating the intention to participate.

The *Exposure* subjects were drawn from the database, which contained all subjects in the base sample. Those subjects, who responded to the short questionnaire and did not refuse or were not excluded from exposure monitoring, were then contacted by telephone to remind about the study and to agree about the exact timing of the measurement period, time and place for meeting, driving instructions, etc. To start the measurements a junior researcher went to the home and - where the employers accepted - to the workplace of the subject, positioned the MEMs, gave the PEM and instructed the subject with regard to the filter change procedure, the core questionnaire and TMAD use. The subject was also offered a GSM telephone for the 48 hours to easily reach the respective junior researcher in case of any problems or questions. At the end of the 48 hour measurement period, while taking the equipment and the paper material back, the subject was interviewed if there was any problems during the monitoring and the paper material was checked through in case of misunderstandings or unanswered questions and additional questions were asked if needed.

The *Diary* subjects were drawn and contacted similarly, and invited to a meeting where TMADs and questionnaires were distributed and their use was instructed to a small group at a time. Those *Diary* subjects, who could not come to the meetings, were contacted at home or workplace. The TMADs and questionnaires that they completed are almost identical to the *Exposure* subjects'. The *Diary* subjects returned these materials in prepaid and addressed envelopes. The diary subjects could use telephone consultant during the 48 hour diary period and they were contacted again by telephone after they returned the paper material if misunderstandings or unanswered questions were noticed.

2.10. Quality Assurance

The performance criteria of the quality assurance program in *EXPOLIS* were in general to minimize any differences between the Centres which would affect the comparability of the results, and specifically to ensure quantified data for all $PM_{2.5}$, CO and NO_2 exposures and microenvironmental concentrations. A maximum detection limit of 1 $\mu g/m^3$ was requested for all VOCs in the target compound list (see Table 2.3.-1.).

The first performance criteria were pursued by using identical sampling equipment, questionnaires, time-activity diaries and work procedures in all Centres (except for VOCs in Basel), by training the junior researchers together in common workshops and by encouraging daily communication between them between the workshops.

The QA was based on the principles that (i) all procedures must be carefully planned, tested and performed according to standard operating procedures (SOPs) approved by the study director, (ii) each unit of data must be traceable as to who produced it, when, with what equipment and according to which SOP(s), and (iii) if any deviations or irregularities occur they must be recorded. Before the pilot phase, the preparation work for preliminary standard operating procedures (pSOP) was distributed among the Centres and individual researchers. A SOP for preparation of SOPs was applied for guidance as to the structure and contents of each pSOP. Preliminary SOPs were prepared for the pilot stage for all subject, field and laboratory procedures, accepted by the local principal investigators, and distributed to all *EXPOLIS* Centres. They covered contacting and instructing the subjects, the use, maintenance and calibration of the measuring equipment, preparation and positioning of the samplers, collection, identification and handling of the samples, collection and handling of data, as well as

recording and archiving of all data from the study. These pSOPs were tested in the pilot phase, corrected and retested until they were approved as real SOPs for the field work by the study coordinator and the KTL QA Unit. The approved SOPs were distributed to all *EXPOLIS* Centres and filed by the coordinator. Table 2.10.-1. lists the standard operating procedures of the *EXPOLIS* study. The QA Unit also monitored the study in the different *EXPOLIS* Centres according to the study protocol and the respective SOPs. The QA Unit inspected the protocol, and the critical phases of the study in the different *EXPOLIS* Centres to ensure that the research plan and SOPs were followed, and reviews the reports and report to the study director.

Table 2.2.-1. Target sizes for the base population samples, *Exposure* samples and *Diary* samples in each *EXPOLIS* center.

EXPOLIS Center	Primary sample	*Exposure* sample	*Diary* sample
Athens	2.000	50	50
Basel	3.000	50	250
Grenoble	-	81	11
Helsinki	2.523	240	240
Milan	3.000	50	250
Prague	-	50	50
Total	10.523	521	851

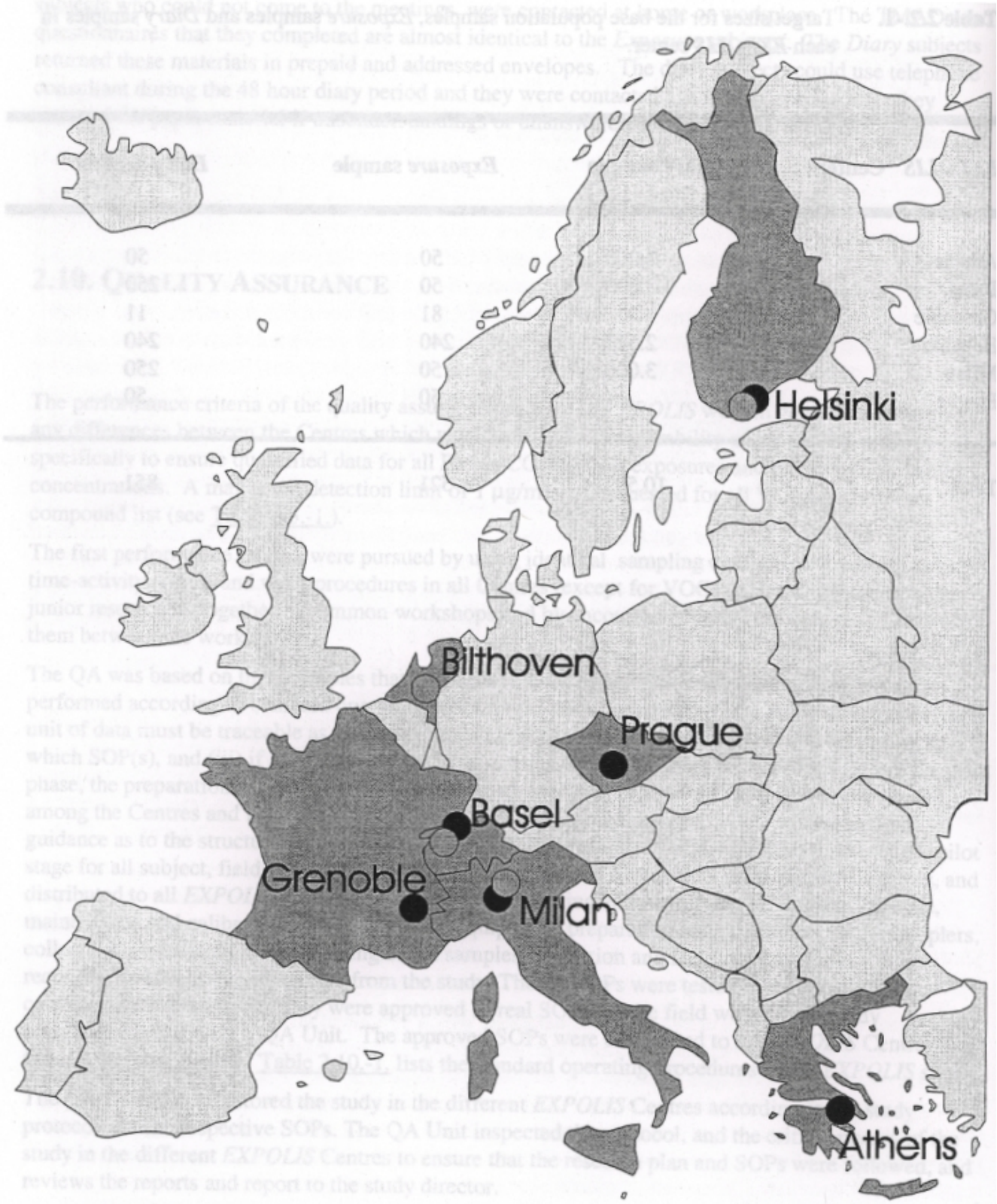

Figure 2.2.-1. The study locations. The dark (red) dots mark the exposure measurement cities. The light (green) dots mark the sample analysis (VTT, Espoo, Finland, Carbotech, Basel, Switzerland, EC/JRC Ispra, Italy) and data analysis (RIVM, Bilthoven, the Netherlands) locations,

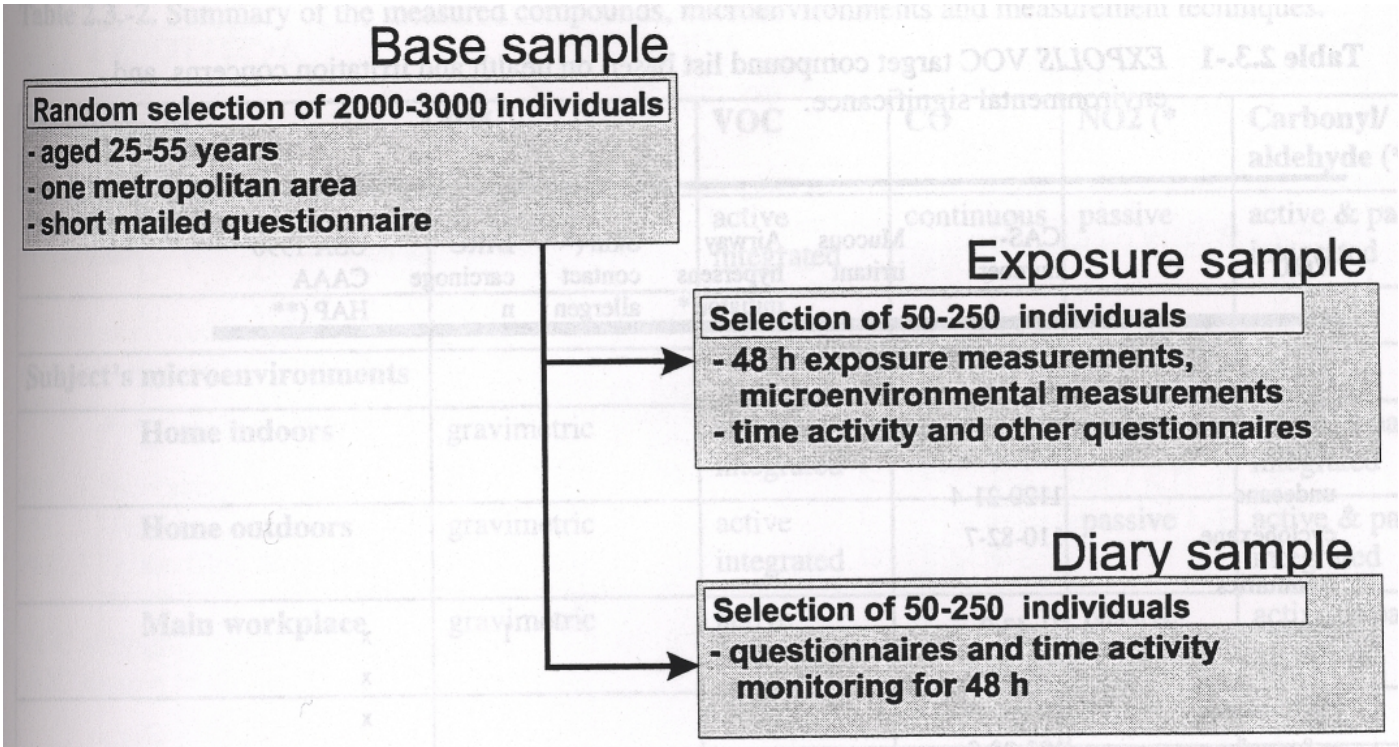

Figure 2.2.-2. Population sampling scheme in each center (with the exception of Grenoble, see text)

Table 2.3.-1 *EXPOLIS* VOC target compound list based on health and irritation concerns, and environmental significance.

VOC	CAS-number	Mucous irritant	Airway hypersens initiator(*	Skin (* contact allergen	IARC carcinogen	USA 1990 CAAA HAP (**
Alkanes						
nonane	111-84-2					
decane	124-18-5					
undecane	1120-21-4					
cyclohexane	110-82-7					
Aromatics						
benzene	71-43-2				I	x
toluene	108-88-3					x
ethylbenzene	100-41-4					x
m&p-xylene	108-38-3					x
o-xylene	95-47-6					x
styrene	100-42-5	Yes	II B		II B	x
naphtalene	91-20-3					x
propylbenzene	103-65-1					
trimethylbenzenes	95-63-6					
Alcohols						
2-methyl-1-propanol	78-83-1					
1-butanol	71-42-0					
2-ethylhexanol	104-76-7	Yes				
phenol	108-95-2	Yes				x
1-octanol	111-87-5					
Esters						
2-buthoxyethanol	111-76-2	Yes		III		
Alkanals						
hexanal	66-25-1	Yes				
benzaldehyde	100-52-7	Yes				
octanal	124-13-0					
Halogenated						
trichloroethene	79-01-6					x
tetrachloroethene	127-18-4					x
1,1,2-trichloroethane	79-00-5					x
Miscellaneous						
d-limonene	138-86-3			II B		
1-methyl-2-pyrrolidinone	872-50-4					
3-carene	13466-78-9					
alpha-pinene	80-56-8					

*) NKB (1994), **) Clean Air Act Amendments of 1990 (1990)

Table **2.3.-2.** Summary of the measured compounds, microenvironments and measurement techniques.

Measurement	$PM_{2.5}$	VOC	CO	NO2 (*	Carbonyl/ aldehyde (*
Personal exposures	gravimetric	active integrated	continuous	passive	active & passiv integrated
Subject's microenvironments					
Home indoors	gravimetric	active integrated		passive	active & passiv integrated
Home outdoors	gravimetric	active integrated		passive	active & passiv integrated
Main workplace	gravimetric	active integrated		passive	active & passiv integrated
Public microenvironments	gravimetric + optical continuous	active integrated	continuous		

*) Additional measurements in some centers only.

Date: _____			LOCATION											ACTIVITIES		
		Briefly	IN TRANSFER					NOT IN TRANSFER						COOK	SMOKING	
		Describe	walk	motor-	car	bus	metro	home		work		other		-	self	same
Time		Activity	bike	cycle	taxi	tram	train	in	out	in	out	in	out	ING		room
8	0		❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑
	15		❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑
	30		❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑
	45		❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑
9	0		❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑
	15		❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑
	30		❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑
	45		❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑
10	0		❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑
	15		❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑
	30		❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑
	45		❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑
11	0		❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑
	15		❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑
	30		❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑
	45		❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑
12	0		❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑
	15		❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑
	30		❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑
	45		❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑	❑

Figure 2.4.-1. The first 5 hours of the 48 h time-microenvironment-activity diary (TMAD) for marking each 15 min of the day at the appropriate microenvironment-activity category/ies. Multiple entries are accepted for each 15 minutes.

Table 2.10.-1. The common standard operating procedures developed for the *EXPOLIS* study.

Preparation of standard operating procedures (SOPs)	SOP Expolis / KTL-G-1.0
Customer procedure, *Exposure* subjects	SOP Expolis / KTL-I-1.0
Customer procedure, *Diary* subjects	SOP Expolis / KTL-I-2.0
MEM sampler positioning and PEM sampler carrying instructions	SOP Expolis / KTL-F-1.0
PM2.5 PEM sampling	SOP Expolis / KTL-F-1.0
PM2.5 MEM sampling	SOP Expolis / KTL-F-3.0
PM2.5 Teflon filter analysis	SOP Expolis / ETHZ-L-5.0
VOC sampling	SOP Expolis / VTT-F-4.0
VOC sample analysis	SOP Expolis / VTT-L-3.0
CO monitoring	SOP Expolis / KTL-F-5.0
Core questionnaire & application	SOP Expolis / UoA-I-3.0
Time-location-activity diary (TMAD) & application	SOP Expolis / UoA-I-4.0

3. METHODS

3.1. Population Sampling

While personal exposure and microenvironmental sampling/monitoring is labourious, questionnaire and time-microenvironment-activity diary (TMAD) application is much simpler. These two methods for acquiring personal exposure information can be combined by sample pooling; drawing *one subsample* for exposure and microenvironmental monitoring plus TMAD and questionnaire application (direct exposure monitoring sample or *Exposure* sample for short), and *another subsample* for TMAD and questionnaire application without exposure or microenvironmental monitoring (indirect exposure assessment sample or *Diary* sample for short). As the unit costs of the *Diary* sample are much lower than those of the *Exposure* sample, a pooled sample may give a smaller variance for population exposure estimate than an *Exposure* sample for the same investment, and the division between the two subsamples can be optimized (Duan and Mage, 1993). Because numerous different pollutants with different costs and presumably different correlations between modelled and monitored exposures were sampled in *EXPOLIS*, no one optimum could be determined for the division between the *Exposure* and *Diary* subsamples.

EXPOLIS includes a large population in only one city (Helsinki) whereas the other Centres had a smaller *Exposure* sample to participate in the full assessment, and a larger *Diary* sample to contribute time-microenvironment-activity diary and questionnaire data only. Thus in one Centre, Helsinki, the aim was to estimate both population exposure distributions and exposure differences between different subpopulations as well as the relative roles of different determinants of exposure, and 240 subjects were drawn for the *Exposure* sample. In the other Centres, the aim was to estimate population exposure levels and distributions for comparison between the Centres and combined analysis of pooled data. The *Exposure* samples consisted of 50 subjects in the other Centres. In addition, samples of another 50 - 250 subjects, depending on the sampling logistics in each Centre, formed the less labourious *Diary* samples. Grenoble was an exception from this design, as described later.

In **Helsinki**, a base sample of the target population was formed by a random draw of 2523 adults (25 - 55 years of age) of the Helsinki Metropolitan Area from the population census. The base sample represents very well the same aged population of the area. A short screening questionnaire about home/work environment, occupation, socioeconomic status, commuting, some personal characteristics and willingness to participate in the study was mailed to this primary population sample. After a second reminder mailing and a computer assisted telephone interviewing, which was done in the Occupational Health Institute in Kuopio, a response rate of 75% was aimed at, and 1881 subjects did return the completed short questionnaire. The final *Exposure* and *Diary* subsamples were drawn at random from the base sample subjects. Those subjects, who had answered the short questionnaire, after having excluded the clearly unwilling or unqualified (e.g. work outside of the area) individuals, were contacted. Similar procedures were applied in other *EXPOLIS* Centres.

In **Athens**, a 2,000 individual random sample of the working age (22 to 55 years old) inhabitants from the entire metropolitan area was formed. A private company was employed to find these 2,000 individuals, visit their homes and complete a short questionnaire with information about their occupational, marital, educational and socioeconomic status as well as home environment and some personal characteristics (age, sex, smoker/non-smoker).

Out of these 2,000 individuals, the final subsamples of *Exposure* (n=50) and *Diary* (n=50) subjects were formed. For the *Exposure* group only non smokers (n=958) were selected. Individuals were selected randomly and contacted by phone to be asked to participate in the study. If they refused, the subject with the next random number was contacted by phone. The procedure continued until an

individual would accept to participate. Approximately one out of every 8 to 10 phone calls was successful.

For the *Diary* group both smokers and non smokers were used. Initially, potential subjects were contacted by mailing. Letters, containing a questionnaire, a TMAD, instructions, a letter from our lab explaining the purpose of the study and a pre-paid envelope to mail back the completed material were send out at given time intervals, approximately 200 at a time. The response rate was in the order of 3 out of 200, so when all 1950 subjects had been contacted only 30 had responded. Consequently, for the remaining 20 subjects, subjects were chosen randomly and contacted by home visits.

In **Milan** the *Diary sample* was based on a random draw from the city inhabitants; the *Exposure sample* was selected from office workers of public and private buildings located in Milan.

For the *Diary sample* (250 subjects) a base sample of the target population was formed by a random draw of 3009 adults (25 - 55 years old) obtained from the Municipal Civil Register of Milan in the *EXPOLIS* database by another researcher. Since in Milan over 75% of the working population operates in offices or similar microenvironments, it was decided to evaluate the exposure for only this category of workers. A short questionnaire was mailed to this primary population sample; it considered occupation, home environment, commuting, some personal characteristics and willingness to participate to the study. The final DIARY group was drawn randomly from the base sample after having excluded the unwilling or unqualified individuals. Each selected subject was personally contacted, about 20 subjects per month, from June '97 to May '98. All the subjects were contacted by the same researcher; they were first contacted by telephone and then visited at their home or workplace for explanations to fill in the TAD and the questionnaires. The subjects were invited to mail the filled questionnaires back as soon as possible by using a return envelope. If a subject had not sent the filled questionnaires back within two weeks, she/he was called and asked for it. When getting back the filled questionnaires, they were checked and if there was something unclear or missing the subjects were contacted again to make the needed corrections. Finally the data were recorded in the *EXPOLIS* database by another researcher.

The *Exposure sample* (50 subjects) was selected from office workers of public and private buildings located in Milan. These buildings had been previously evaluated by our Institute in former studies and can be considered representative of the different building typologies. Fifty office workers were selected among the workers of these buildings and the adopted criteria were: age (15 - 55 years of age), place of residence (Milan) and job (only office workers). The selected subjects were informed on the aims and the methodology of the study. The measurements were performed from March ‘97 to January '98 (about 6 subjects per month) following the *EXPOLIS* standardized procedures.

In **Basel,** a random sample of 3'000 persons (25-55 years, male 50.5%, Swiss 68.6%) has been drawn from the local civil register. A short screening questionnaire on socio-economic status, home environment and willingness to participate has been mailed to this base sample which represents very well the target population of *EXPOLIS*. A total of 1862 subjects (62.1%) returned the short questionnaire; 626 (33.6%) immediately after receiving the questionnaire, 928 (49.8%) after a first recall and 308 (16.6%) after a second recall. In total, 404 subjects had to be excluded, mostly because they had sent back an empty screening questionnaire, resulting in a base sample of 1458 subjects (48.6%) with valid short screening questionnaires. From the 557 subjects (18.6%) willing to participate, the *Exposure* and *Diary* sub-samples were drawn at random and contacted by phone to be asked to participate in the study. If they refused (31 subjects for the diary group and 11 for the exposure group) or could not be reached after several trials, the subject with the next random number was contacted. From the 328 subjects who were recruited for the *Diary sample* and instructed in groups of 5-15 persons, 272 sent back valid core questionnaires and TMAD’s, 10 sent back only a valid core questionnaire. All 50 subjects who were enrolled for the *Exposure sample* completed the *EXPOLIS* protocol.

In **Prague** the base sample was also based on a random draw from the city inhabitants. However, - like in Milan - the *Exposure* and *Diary* samples were drawn from the municipality employees. These

samples will be compared to the larger primary population samples, and the results will be statistically corrected as necessary to represent the more general population.

In **Grenoble** an ongoing study on the $PM_{2.5}$ exposures and daily symptoms of 40 volunteers (20 asthmatics and 20 controls) 20 to 60 years of age, was adapted to yield $PM_{2.5}$ exposure results which can be related to the data from other *EXPOLIS* Centres. Contrary to Helsinki and to the other *EXPOLIS* Centres, Grenoble only studied one main *Exposure* sample. The *Diary* sample consisted only in 11 further volunteers. The *Exposure* sample was studied in 2 phases : a summer phase (phase 1 : May - July 1996) and a winter phase (phase 2 : January - March 1997). The phase 1 *Exposure* sample consisted in 40 volunteers. The phase 2 *Exposure* sample consisted in 41 volunteers, plus the 11 *Diary* volunteers. Among the phase 2 *Exposure* sample, 27 volunteers already participated phase 1. So, the total Grenoble *Exposure* sample consisted in 81 measured volunteers including 27 for whom we have repeated (2) measurements plus 27 persons for whom we collected data only once (either during phase 1 or 2).

Volunteers were recruited with the help of the Grenoble Hospital pneumology service and using a public appeal in a local newspaper: half of the volunteers were asthmatics, half were controls (20-60 years old). They were living and working in Metropolitan Grenoble. The short questionnaire was filled in by 40 *Exposure* persons for phase 1 and 41 for phase 2. The long questionnaire was filled in by 39 and 41 volunteers for phase 1 and 2 respectively. The *Diary* sample filled in 11 short questionnaires and 7 long questionnaires only during phase 2. The original English questionnaires were translated and back-translated in french.

Results of the population sampling process and descriptions of the primary population samples and the *Exposure* and *Diary* samples will be published separately. The primary, *Exposure* and *Diary* sample sizes in each *EXPOLIS* centre are listed in Table 2.2.-1.

A data integrity protocol was established according to the data security requirements of the *EU Directive on Protection of Individuals with Regard to Processing Personal Data in Medical and Epidemiological Research*. This protocol includes the contents and security of the *EXPOLIS* databases, use of person code numbers which cannot be translated back to identity, and training for the whole staff.

3.2. Questionnaires and Time-activity monitoring

EXPOLIS used four questionnaire-based data collection tools:

1) Short Screening Questionnaire,

2) Core Questionnaire,

3) Time-Microenvironment-Activity-Diary (TMAD, Figure 2.4.1), and

4) Retrospective 48 h Exposure Questionnaire.

The English versions of questionnaires 1, 2, and 4 can be found in Annex I.

The purpose of the *Short Screening Questionnaire* was particularly to evaluate the subjects' intention for participation and to get some background information about them. In Helsinki and Basel some information was directly obtained from the census. In Helsinki: gender, birth year, home type, home area, number of adults in the house and number of children in the house; in Basel: gender, birthday, nationality. This information was collected with the short questionnaire in other Centres. Because of different time of delivery of the population sample in each Centre, the mailing of the short screening questionnaire took place at different moments. The short screening questionnaire changed a little from the first Helsinki mailing till the last central Europe mailings. The main difference between the Short questionnaire versions are, that in Helsinki we asked the subject's occupation and the other Centres asked years of education instead to define the socioeconomic status of the subject (The years of education is asked from the exposure and diary samples in Helsinki.). The short questionnaire answers were checked and updated from exposure and diary samples during the measurement. See Annex I and Tables 5.1.2/A...P and 5.1.3 about the short screening questionnaire data.

The *Core Questionnaire* covered the indoor air quality related characteristics of each subject's home and workplace, as well as commuting and some exposure related personal characteristics, such as smoking. The study subjects filled in the core questionnaire by themselves in their own language. For most of the questions they chose the answer from the given alternatives. All the subjects were given both written and oral instructions how to fill in the questionnaires. See Annex I and Tables 5.2.1/A ... 5.2.5./P about the core questionnaire data.

The *TMAD* was needed to assess the times that subjects spent in each microenvironment and activity while their personal exposures and the microenvironmental concentrations were measured. Usually the diary was collected e.g. from Monday morning to Wednesday morning (6 - 6 A.M.) or from Wednesday evening to Friday evening (6 - 6 P.M.) for 48 hours, but sometimes some hours were missing or some extra hours were measured because of shorter or longer measurement period (time of picking up the equipment varied). The TMAD asked the subjects to mark each 15 min of the day at the appropriate microenvironment-activity category (see Figure 2.4.-1.). The microenvironment categories in this TMAD are *in transfer* (*walk/bike, motor cycle, car/taxi, bus/tram,* and *metro/train*) and *not in transfer* (*home in* and *out, work in* and *out, other in* and *out*), and activities are *cooking, smoking self* and *smoking in same room*. Multiple entries (e.g. home indoor, home outdoor, car) are allowed for each 15 min. In the analysis each 15 min is divided evenly between all entries. Instructions how to fill in the diary were given both in written format and orally. See Figure 2.4.-1. and Tables 5.4.1./A ... 5.4.3./P about the Time activity diary.

The *Retrospective Short-Term Recall Questionnaire* referred to 'the last 48 hours' and was requested to be completed at the end of the 48-hour PEM/MEM measurement period of each subject. The 48-hour recall questions addressed specific activities which may influence personal exposure, particularly to VOCs (cleaning, gluing, etc..). Subjects were also asked to what extent (0-10 continuous scale) they were annoyed from air pollution during the measurements in their homes, at their workplaces and in traffic and what was the main cause of annoyance. Furthermore, the exposure sample was asked whether they kept the PEM-case near them for all of the measurement period and if not, what periods they did not have it with them. See Annex I and Tables 5.3.1./A ... 5.3.4. about the Retrospective Short-Term Recall Questionnaire.

All questionnaires were originally prepared in English, and translated to the 6 *EXPOLIS* languages and back-translated independently to control for a common meaning and understanding of each question. As some TMAD locations (e.g. metro in Basel) and some questions (e.g. gas fired hot water heaters in Helsinki) are irrelevant in some Centres, such locations and questions were omitted in local translations. Centres also added some questions of local research interest to the questionnaire.

However, for the need of combining the data the coding of the common locations and questions remained the same in all Centres. After the monitoring period the paper material were checked by the junior researcher and discussed with the subject.

3.3. PM2.5 Sampling and Analyses

3.3.1. Methods

Personal Exposure Sampling

The $PM_{2.5}$ exposure was measured by a personal exposure monitor (PEM) (Figure 3.3.-1.). The sampler was carried by each subject for a sampling period of 48 hours. It consisted of a pump, a cyclone and a filter holder packed together with a Langan CO monitor, VOC sampling tube and battery pack into a rigid aluminum briefcase which was filled with noise absorbing material. The modified Buck IH pump (A.P.Buck Inc. Orlando, FL, USA) was silent, lightweight, capable for sampling over 48 h with six D-size alkaline batteries and was therefore suitable for personal measurements. It draws air at 4 L/min using a simple volumetric (it keeps the pump speed constant) flow control. Small $PM_{2.5}$ cyclones (GK2.05) were designed and constructed for *EXPOLIS* by BGI Inc. (Waltham, MA, USA). The design and the performance of the GK cyclones are presented in Kenny and Gussman.(1997) With this cyclone design the filters can be handled from pre to post weighing in 37 mm Millipore filter holders (Millipore Corporation, Bedford, MA, USA) which decreases the risk of filter contamination and damage. Gelman Teflo filter (2 Φm pore size) was chosen for the sampling medium, because it has a high collection efficiency, a low pressure drop and a low chemical background.

In the laboratory the flow rate was adjusted to 4 l/min with a bubble flow metre (e.g. Buck M-30 by A.P.Buck Inc. Orlando, FL, USA) before and controlled after each 48 h sampling period, with the cyclone and the actually used filter in the sampling line.

Two filter holders were provided for each subject. One 'day filter' for the two sampling periods beginning at leaving home for work and ending at returning home from work (about 2 x 9-10 hours), and one 'night filter' for the remaining times (about 2 x 14-15 hours). The subjects were instructed on how to change the (day or night) filter holders by simply pulling the first filter holder apart from the cyclone and pressing the second filter holder in its place.

Microenvironmental Sampling

Microenvironmental monitors (MEMs) (Figure 3.3.-2.) were placed at the subject's home (indoors and outdoors) and workplace for 48 h to collect microenvironmental $PM_{2.5}$. The pumps were programmed to run at home during the expected non-working hours and in the workplace during the expected working hours of each subject. The flow rate was measured and adjusted before and controlled after each sampling with a bubble flow metre (e.g. Buck M-30).

The MEM sampler contained an EPA-WINS impactor (EPA Well Impactor Ninety-Six, BGI), a 47 mm filter holder (BGI) with a Gelman Teflo filter and a PQ100 pump (BGI). A Graseby-Andersen PM_{10} inlet was used in outdoor measurements to avoid wind and rain effects. The EPA-WINS is a single jet well impactor designed to remove particles with a 50% cut size at 2.5 μm particle diameter at

16.7 l/min (USEPA 1997, RTI 1996).

The PQ100 pump was equipped with a microprocessor controlled timing and mass flow adjustment system. It runs from mains power (220/110VAC) or a 6 V internal lead acid battery (up to 32 h) and it was enclosed in a weatherproof case. The pump is designed to pull in a sample of air at a constant mass flow controlled rate of 1.0 - 25 l/min (± 5 %). Aluminium Y-joints for two filter holders were prepared for an option (e.g. a blank filter and an exposed one, or for two filters in rooms with heavy smoking).

Air flow:

Because the PEM pump has a volumetric flow control and the MEM pump a mass flow control, the air flows had to be normalised by air pressure and temperature. Normalisation is particularly important in the cold Nordic winter (Helsinki) when the temperature difference between indoor and outdoor air is 20-50 °C.

In *EXPOLIS* all sample volumes were normalised to 101.3 kPa (760 mmHg) air pressure at 20 °C. The pressure data for PEM flow normalisation were taken from a fixed meteorological station. The PEM temperature results were normalised using the temperature data collected by the external temperature sensor of the Langan CO monitor. The MEM adjusted the drawn air mass to be equivalent to 16.7 l/min at 20°C and 101.3 kPa automatically.

Gravimetric Analysis

When a microgram sensitivity is needed in the weighing, a micro balance placed on the stable stone table in a mechanically and physically stable room must be used. The Teflon filters are electrically non-conductive and will hold the static charges collected on them. They can be deionised by an alpha radiation source (Po-210), an ionising bar or a piezoelectric crystal gun. If this problem is not controlled, repeatable weighing results are impossible.

The filters were weighed in each *EXPOLIS* Centre by their own equipment. Microbalances (e.g. in Helsinki Mettler MT5 by Mettler-Toledo AG, Greifensee, Switzerland) were used for weighing the filters. Each filter was stabilised in the weighing room for a minimum of 16 hours prior to weighing. After the sampling the time of stabilisation was limited to a maximum of 36 hours.

The weighing conditions were controlled by recording the temperature, relative humidity and air pressure before and after each session. The standard weight was also weighed before and after each session, and each filter was deionised at both sides with a Po-210 deioniser (Staticmaster 1269 by Cahn Inc. USA) or a Multistat deioniser (Haug Biel Gmbh, Germany) before each weighing. Two consecutive weighings within 1 Φg were required before a mass for a filter or the standard weight was accepted and recorded.

When the weighed net mass (and volume) is small (less than 1/1000) compared to the filter mass (and volume), air buoyancy becomes an important issue and the mass results must be corrected for any changes in air density between the pre- and post- sample weighing. This is the case for example when weighing fine particle samples by personal or microenvironmental monitors.

Because the effect of air buoyancy depends on the air density i.e. air pressure, temperature and relative humidity, when weighing the filters, all masses in *EXPOLIS* were corrected according to equation 1:

$$\Delta b = V_f H(\rho_{a2} - \rho_{a1}) H10^{9} \quad (1)$$

where: Δb = buoyancy corrected net mass (:g)

V_f = filter volume (m^3)

ρ_{a2} = air density in the post weighing conditions (kg/m^3)

ρ_{a1} = air density in the pre weighing conditions (kg/m^3)

Air density was calculated according to equation 2 (Mettler Toledo 1994):

$$\rho_a = \frac{3.484\,HP - (0.00252\,HT - 0.02058)\,HR.H.}{273.2\,HT} \quad (2)$$

where: ρ_a = air density (kg/m^3)
P = atmospheric pressure (hPa)
T = temperature (EC)
R.H. = relative atmospheric humidity (%)

Buoyancy correction for Mettler MT5 balance was calculated according to equation 3 (Mettler Toledo 1994)

$$m = R\,H\,\frac{1 - (\rho_a / \rho_w)}{1 - (\rho_a / \rho_f)} \quad (3)$$

where: m = mass (g)
R = balance display (g)
ρ_a = air density (kg/m^3)
ρ_w = calibration weight density = 8000 (kg/m^3)
ρ_f = density of weighing sample (here a filter) (kg/m^3)

The correction according to equation 4 for Mettler MT5 results is needed because of the difference between the filter density and the calibration weight density (8000 kg/m^3). No correction is needed for steel (= 8000 kg/m^3). In general, the need for a buoyancy correction depends on the balance type and its internal design used for correcting the buoyancy effect.

In the weighing room climate the most powerful factor for the buoyancy (i.e. air density) change is air pressure. The effects of temperature and relative humidity are much weaker. An example in Table 3.3.-1. highlights the magnitude of change in the observed (uncorrected) mass reading when the conditions change by +30 mmHg, +2 °C or +20 % RH between pre and after weighing of a filter. If all the factors work towards the same direction e.g. increase the observed mass, the total effect of the climate change in the weighing room could be up to 10-15 μg.

It must be underlined here that for a change in relative humidity this correction only applies for its effect of air buoyancy. Filter mass may also need correction for relative humidity changes because of possible mass changes in hygroscopic PM collected on the filter and also for the hygroscopicity of the filter itself. The need to correct the effects of hygroscopicity will be evaluated and applied as necessary after the field work. However, based on hygroscopicity of sulphate, the most abundant hygroscopic compounds in most $PM_{2.5}$ samples, the hygroscopicity of the sampled mass should not be significant

below RH 65 % (Tang 1980).

QA/QC

Standard Operating Procedures (SOPs):

To assure the quality and comparability of the data collected in the different *EXPOLIS* Centres SOPs were developed and used for all laboratory, field and subject procedures. The SOPs of the U.S.EPA National Human Exposure Assessment Survey (NHEXAS) (Lebowitz *et al.* 1995) and the Good Laboratory Practice (GLP) regulations of OECD (OECD 1992) as applied in National Public Health Institute of Finland (KTL) Division of Environmental Health were used as models to develop the SOPs for *EXPOLIS*. The Quality Assurance Unit of KTL - Environmental Health supervised all procedures in this study.

Duplicates and blanks:

Duplicates and field blanks were measured to assess the repeatability and detection limits of the method and possible filter contamination. The duplicate and field blank filters were included in the measurements throughout the whole field work duration and they were distributed equally between all the microenvironments (home indoor, home outdoor and work). The number of field blank and duplicate filters was set at 5 % of the number of actual sample filters but no less than 20 filters to assure statistical usability of the results. The average field blank $PM_{2.5}$ mass increase was subtracted from all the exposed $PM_{2.5}$ mass results.

The duplicates were measured using identical monitors sampling side by side. The PEM samplers were either carried by the researchers, placed in one location, or included in separate microenvironmental measurements done e.g. in transportation vehicles nominally for 24 hours. The MEM duplicates were nominally sampled for about 30 hours at home (indoors and outdoors) and about 15 hours at the work place, depending on the actual schedule of the subject being monitored.

The blank filters used in *EXPOLIS* underwent all procedures in the lab and the field except that no air was drawn through them. The PEM field blanks were placed in the sampling case, in their filter holders with the protective plugs closed for the whole sampling period of about 48 hours. The MEM field blanks were applied by using an aluminium Y-joint for 48 hours. Its one end was connected to the EPA-WINS impactor and the other ends to two filter holders. Both filter holders were equipped with a filter but air was drawn only through one.

Weighing tests:

Because the flow rate of PEM was only 4 l/min and the shortest sampling periods only 16 hours, low masses (the lowest with PEM day filters, typically 30-50 μg) could be expected. For that reason the weighing errors had to be minimised. For this purpose, three pre field tests were carried out. The effect of static charge on the repeatability of the results was tested. In the first test five (37 mm Gelman Teflo) filters were weighed 10 times each in a Faraday cage, which is an option in the Mettler MT5 microbalance. Faraday cage eliminates the static charge effects to the weighing results. Also a 200 mg stainless steel standard weight was measured 17 times with a Faraday cage. The second test was carried out using the small weighing pan which is normally used in this type of balance. In this test five filters were weighed five times each and the standard weight was weighed 25 times. The third test was also carried out with the small pan, but both sides of the filters were deionised with a Po-210 deioniser before each weighing.

PEM-MEM Comparison:

Because two different types of $PM_{2.5}$ samplers were used in PEM (cyclone) and MEM (impactor) sampling, these two methods had to be tested side by side to evaluate PEM and MEM data comparability. This parallel $PM_{2.5}$ sampling test was run for 45 h inside a laboratory in the city centre of Helsinki. In this 'Helsinki test' five PEMs, five basic indoor MEMs, three indoor MEMs equipped with a Y-joint, and two outdoor MEMs with a Graseby-Andersen preimpactor were tested in parallel to compare the 4 l/min cyclone based PEM sampler with a 16.7 l/min impactor based MEM sampler.

Additional PEM-MEM comparison tests were carried out indoors and outdoors in Helsinki and Basle. In this 'multicentre test' PEMs and MEMs were set side by side into a sampling site to test the comparability in different circumstances and concentration levels. The sampling duration was 24 hours in all but one of these tests, where it was 46 hours.

3.3.2. Results

Pilot

Weighing repeatability for blank filters was poor in the beginning. The main reason proved to be the static charge in the filters. After getting the Po-210 deioniser and using it before each weighing, the repeatability problem could be solved.

PEM characteristics like weight, portability, noise level and capability to run 48 hours with batteries and overall reliability were tested and proved to be acceptable for measuring personal exposures according to procedures described in the SOPs.

In the first PEM-MEM comparison tests PEM results were often found to be smaller than MEM results. This problem had to be solved before the field phase. In the PEM filter holder a leakage between the parts of the plastic filter holder was found and consequently a fraction of the air flow bypassed the filter. This could be observed by sampling the filtered air with an optical particle counter. The fitting joint between the bottom and the centre part of the filter holder had to be tightened by compressing the pieces together symmetrically and with considerable force. Also the joint had to be taped to avoid any loosening during the field use, especially by the study subjects. The correct procedure, once tested and understood was easy to learn. After the careful tightening and taping procedure the PEM and the MEM results were comparable.

Also the timing routines in the MEM proved to be incomplete to the *EXPOLIS* procedure of two consecutive sampling periods in the home indoor, outdoor and workplace. The software was then modified by BGI Inc. to meet the *EXPOLIS* needs.

Because of the static charge problem in filter weighing and the PEM filter holder leakage, the mass concentration results for the pilot subjects were not reliable and were not used in any exposure analyses, but only for procedural development.

QA/QC

SOPs:

According to pilot results the procedures were finalised and written into SOPs. The following SOPs were developed for $PM_{2.5}$ sampling:

MEM sampler positioning and PEM sampler carrying SOP describes how the home indoor, home outdoor and workplace MEM samplers should be located in various personal and microenvironmental settings and how the subject should keep the PEM sampler in different circumstances, and how the subject should carry it when moving.

$PM_{2.5}$ PEM Sampling SOP describes the personal $PM_{2.5}$ sampler. It covers the preparation, calibration and use of the PEM sampler, and the field blank and duplicate procedures.

$PM_{2.5}$ MEM sampling (Indoor and Outdoor) SOP describes the particulate sampler for collecting $PM_{2.5}$ in indoor and outdoor microenvironments. It covers preparation, calibration and use of the MEM sampler, collection and handling of the samples and field blank and duplicate procedures.

$PM_{2.5}$ Teflon Filter Analysis SOP describes the handling of the Teflon filters in the laboratory before and after exposure, including coding for sampling, conditioning and weighing before and after sampling, filter holder preparation, used filter storage, data coding and filing and quality control.

Duplicates, blanks and detection limit:

The test results after the pilot and some early field phase duplicate results for PEMs and MEMs in Athens, Basle and Helsinki, are presented in Figures 3.3.-3 and 3.3.-4. The method to analyse and report duplicate results has been presented in an article by Bland and Altman.(1986). The average absolute difference for PEM duplicates was 2.1 μg/m^3 and the standard deviation 2.0 μg/m^3 and for the MEM duplicates 0.7 μg/m^3 and 0.6 μg/m^3, respectively.

The field blank filters showed some systematic mass increase during the field measurements. The average mass increase and standard deviation in Helsinki for MEMs (n=74) were 5.8 μg and 5.1 μg and for PEMs (n=66) 1.8 μg and 4.1 μg, respectively.

The detection limit was defined as three times the standard deviation in field blanks divided by the sampled volume (16 m^3 for MEM and 4.3 m^3 for PEM). The detection limits were 1.0 μg/m^3 for MEM and 2.8 μg/m^3 for PEM.

Filter Weighing:

The three weighing tests show that reducing the effect of static charge of the filters by the manufacturer's Faraday cage did not produce satisfactory weighing precision probably because of increased instability of the weighing system (Table 3.3.-2). When using the standard weighing pan without deioniser, the weighing precision was unacceptable for the filters - due to electricity - but good for the standard weight. The wanted results were achieved when the standard weighing pan was used together with careful deionisation. The standard deviation of the filters was then 1.7 μg, which is almost as good as for the standard weight with the pan.

In Helsinki the data quality was improved by auto loading the weighing results directly from the balance to a computer using a data transfer software (Balance Link Ver. 2.20) developed by the balance manufacturer (Mettler-Toledo AG, Switzerland) to eliminate data entry errors.

PEM-MEM Comparison:

In the side by side 'Helsinki test' for all PEM and MEM samplers (N=15) the average observed indoor air $PM_{2.5}$ concentration was 7.1 μg/m^3 (RSD=3.0%) (Figure 3.3.-5.). For the 5 PEMs the result was 7.2 μg/m^3 (RSD=2.6 %) and for the 10 MEMs 7.0 μg/m^3 (RSD=3.1%). Results of the 'inter centre test' of the personal GK2.05 cyclone (PEM) and the EPA-WINS impactor (MEM) in Helsinki and Basle are presented in Figure 3.3.-6. The mean MEM/PEM ratio in this test was 1.04. If two outliers, the two highest concentration points, are left out from the analysis the MEM/PEM ratio was 1.02.

3.3.3. Discussion

The methods used in *EXPOLIS* to determine the human exposures to and microenvironmental concentrations of $PM_{2.5}$, and the quality assurance protocol were developed especially for this project. The methods were proven to be reliable and applicable for measuring exposures down to quite low $PM_{2.5}$ concentrations in the different *EXPOLIS* Centres.

Pilot

Pilot phase proved to be necessary for finalising the equipment and operating procedures for the *EXPOLIS* field work. The most serious problems, which were solved in the pilot, were the portability of the PEM case and its noise prevention. Noise problem was caused by the air pump in the *EXPOLIS* PEM case. It could be solved by adding noise prevention material, but at the same time the weight of the case increased. Optimisation between these two problems was accomplished in the pilot tests. The PEM filter holder leakage was also found and solved in the pilot tests. After changing the procedure the study subjects changed the filters without sample losses.

While weighing low net masses, minimising and correcting for the weighing errors becomes a critical issue. This was achieved by first deionising the filters carefully at both sides just before weighing, correcting the observed filter masses for the buoyancy differences due to changing air density and ensuring that relative humidity remained below 65 % in filter conditioning and weighing room. According to air buoyancy calculations, it is obvious that better weighing accuracy can be achieved by correcting the effect of air buoyancy mathematically, using non-hygroscopic filters and keeping the weighing room relative humidity below 65% RH, than by using the typical climate controlled room without air buoyancy correction. This observation can save considerable amounts of money when building and operating weighing rooms.

Buoyancy correction is hardly ever reported in the published papers. This correction becomes increasingly important in the future as more studies will be carried out for fine ($PM_{2.5}$) and ultra fine (PM_1) particles with particularly low sample to filter mass ratio in personal exposure studies.

QA/QC

SOPs:

Detailed Standard Operating Procedures are necessary in all field studies with a number of staff members conducting repeated tasks. SOPs are even more essential in multicentre and multinational studies to ensure data quality comparability.

Duplicates and blanks:

PEM duplicate results (Figure 3.3.-3.) show, as expected, larger average difference and standard deviation than MEM duplicates, but in both cases the differences are small. This could be due to the smaller sampled masses and weaker flow control of the PEM compared to the MEM. It can also be seen that the absolute difference between duplicate samples does not depend on the concentration level or particle composition, which may vary between different cities and microenvironments.

The field blank results show systematic blank filter mass increase, which, is about 20% of the corresponding PTEAM (Thomas *et al.* 1993) and a Dutch study (Jansen *et al.* 1998) results. The reason or contents of this increased mass is not known. One reason might be the time between pre and

post weighing of the filters, which was 9 days on average in *EXPOLIS*, but could be longer in the above mentioned studies.

PEM-MEM Comparison:

Side by side PEM - MEM method comparison in Helsinki (Figure 3.3.-5.) shows that the results are repeatable and there are no systematic differences between the results obtained by the PEM and the MEM sampler with or without Y-joint or outdoor sampling inlet. PEM - MEM comparison test results in Helsinki and Basle (Figure 3.3.-6.) show good agreement between these two methods and thus these monitors and procedures were used in the field work of *EXPOLIS*.

In general the results show that the consistent quality assurance work done at each stage of the $PM_{2.5}$ sampling and filter handling has been necessary and has paid off. Application of the equipment and procedures to *EXPOLIS* field work is likely to yield reliable results.

3.4. VOC:s Sampling and Analysis

3.4.1. Materials and Methods

Target VOC Compounds

VOC compounds measured in *EXPOLIS* were selected on the basis of their environmental and health significance or of the usability of one or group of few compounds as markers of pollution sources. The availability of one single sampling and analysis method for all target compounds was also required. The list of selected VOC target compounds included 30 different compounds and is shown in Table 2.3.-1, including their characteristics and sampling methods applied.

Sampling Media

In Athens, Milan, Prague and Helsinki, VOC's were sampled with stainless steel sampling tubes (dimensions: 90 mm ∗6.35 mm o.d.) (Perkin Elmer) containing approximately 250 mg of Tenax TA adsorbent (60-80 mesh) (Chrompack, Middelburg, Netherlands). In Basle, VOC samples were collected with adsorbent tubes containing two stacks of active charcoal (25 mg each, 0.05-0.1 mm particle size) which were stabilized with 4 silver nets (VDI 3482, Blatt 4 and 5).

Tenax is a widely used adsorbent in VOC sampling (Versino *et al.*, 1974, Pellizzari *et al.*, 1984, Class and Ballschmitter, 1986). It is stable at temperatures up to 250 °C, allowing thermal desorption of sampled compounds. Drawbacks with Tenax include some artefact formation of chemicals such as benzaldehyde, acetophenone (Hutte *et al.*, 1984, Walling *et al.*, 1986, Rothweiler *et al.*, 1991, Cao and Hewitt, 1994) and phenol (Pellizzari *et al.*, 1984) and inability to retain very volatile organic chemicals e.g. low molecular weight aldehydes and ketones, alcohols, and amines (Rothweiler *et al.*, 1991, De Bortoli *et al.*, 1992, Rothweiler *et al.*, 1992). Increasing polarity of the adsorbate also decreases the efficiency of adsorption. This has been demonstrated for acetic acid, isopropanol, and 1,2-ethanediol (Rothweiler *et al.*, 1992).

Tenax TA is a further development of Tenax GC. It can be cleaned simply by heating in an inert gas flow. The background concentrations in cleaned Tenax TA sampling tubes are low (De Bortoli *et al.*, 1992, Brown, 1996) and Tenax TA produces less artefacts than Tenax GC (McLeod and Ames, 1986).

In Basel the VOC samples were collected into active charcoal. With its high adsorbing capacity for

VOC's this is one of the most widely used adsorbents for preconcentration of VOC's in connection with liquid desorption. After sampling, VOC's are desorbed with carbondisulfide from the sampling tube and the eluate is analysed with a gas chromatograph equipped with a suitable tandem capillary column and an ion trap detector. The method is suitable for the measurement of individual VOC's (b.p. 60-320 °C) in non-industrial buildings and outdoors in the concentration range of 0.5-5 $\mu g/m^3$ to 0.1-1 mg/m^3. The method is not suitable for compounds which cannot be desorbed quantitatively with carbon disulfide from charcoal, e.g. naphthalene, styrene, benzaldehyde, hexanal, octanal, 2-buthoxyethanol, 2-ethyl-1-hexanol, 1-methyl-2-pyrrolidinone and the monoterpenes. The background concentration of cleaned active charcoal tubes should not exceed 30 ng/tube for benzene and 45 ng/tube for toluene.

VOC Sampling

Sampling Principles

VOC's were sampled into a sampling tube by the vacuum of the $PM_{2.5}$-pump, which was connected to the VOC sampling line by a T-joint. In Tenax TA sampling, the target sample volume was 2-4 L and the VOC sampling flow was adjusted by an experimentally designed flow-restrictor/diffusion-barrier system made of stainless steel capillary tubes before and after the sampling tube. The target sample volume for active charcoal tubes was 30-50 L and the VOC-sampling flow was adjusted with a valve.

Without any diffusion-barrier system, the tubes would be more or less contaminated by passive diffusion especially in the ultra low flow rate Tenax TA sampling, see Table 3.4.-1.

While the $PM_{2.5}$ filters were loaded with particles during the sampling period, their flow resistance and, consequently, the VOC sampling flow rate increased accordingly, giving systematically increased weight to the VOC concentrations occurring towards the end of the sampling period. In personal exposure measurements, some errors in determining the total sample volume may have occurred, because of the differences of the flow resistance of the ″night” and ″day” filters.

Sampling Monitors

The PEM (VOC sampling tube, sampling pump, cyclone and holders with filters for $PM_{2.5}$, CO monitor with external temperature sensor, and a battery back) (Figure 3.3.-1.) was packed into a 5.2 kg (total) aluminum briefcase. Aluminum was chosen as case material, because it is free of VOC emissions.

MEMs (VOC sampling tube, sampling pump, impactor and filter holder with filter for $PM_{2.5}$) (Figure 3.3.-2.) were packed into a portable sound absorbing container. The MEM container was made of MDF-board coated with a low emission paint. According to tests done by VTT, the VOC-emissions of the container material were low and qualitatively of no interest in relation to the target compound list.

Sampling Procedure

The personal, home-in, home-out and workplace VOC samples were collected for each subject in one VOC sampling tube each. The air flow rates through sampling tubes were measured in the laboratory before and after each subject period with a bubble flow metre (Mini Buck Calibrator M-1, A.P.Buck Inc., Orlando, FL, U.S., or similar).

In PEM measurements with a sampling time of 48 hours, the VOC-flow rate was adjusted to 0.5-1.0 mL/min for Tenax TA and 10-18 mL/min for active charcoal samplers. In PEM measurements, two different $PM_{2.5}$ filters were used (“night” and “day” filter). When the subject’s personal sampling was started in the evening (e.g., 6:00 PM), the VOC flow rate before sampling was measured with the "night" filter and after sampling with the "day" filter connected.

For MEMs, the VOC sampling times were identical to the PM2.5 sampling times; typically 26-30 hours at homes and 14-18 hours at workplaces. For Tenax TA and active charcoal tubes the VOC flow rates were adjusted to 1.5-2.5 mL/min and 16-75 mL/min, respectively. The microenvironmental $PM_{2.5}$ samples were collected on one $PM_{2.5}$ filter each. Thus the VOC-flow rate before and after sampling was measured with the same $PM_{2.5}$ filter installed.

When delivering and collecting back the equipment to/from home indoor and workplace, temperature and RH (Extech 445922, Extech Inc., Waltham, MA, U.S.) were measured. After each measurement all sampling data was recorded on the *EXPOLIS* database.

VOC Sample Analysis

The cleaning conditions of the Tenax TA sampling tubes were selected so, that no degradation of the polymer occurs, e.g., at the temperature of 280-300 °C for at least 2 hours using a gas flow of 30-50 mL/min. Prior to use, the sampling tubes were reconditioned in a thermodesorption device (Perkin

Elmer) at 260°C for 6 minutes. Clean, tightly sealed sampling tubes were stored in a closed, emission free containers at most for two weeks before sampling.

The VOCs samples on Tenax TA were analysed by VTT using Perkin Elmer ATD 400 thermal desorber and Hewlett-Packard 5890 Series II+ gas chromatograph equipped both with a flame ionisation detector (FID) and a mass selective detector (MSD). The methodologies for VOC sample analysis and VOC sampling used in this *EXPOLIS*-study have been described in detail in respective Standard Operation Procedures (VOC-sampling SOP *EXPOLIS*/VTT-F-4.0 and VOC sample analysis SOP *EXPOLIS*/VTT-L-1.0). The VOCs were desorbed from the exposed sampling tubes in Perkin Elmer ATD 400 using the following conditions:

desorption time	6 minutes
desorption temperature	260°C
desorption flow	50 mL/min (Helium)
cold trap temperature	-30°C (filled with Tenax TA)
flash desorption temperature	280°C

VOCs released by heating were transferred to the gas chromatography via heated transfer line. In the gas chromatography the sample was splitted to 1:1 in two identical non-polar separation columns (type PONA, length 50 metres, internal diameter 0,2 mm, phase thickness 0,5µm). One column was connected to the FID and the other respectively to the MSD (Hewlett-Packard MSD 5972), so each Tenax-sample was analysed with both detectors.

Single VOC-compounds in the sample were identified from the MSD total ion chromatogram using Wiley 275 software library. In general, 70 to 95 % of the VOCs were identified. The 30 target compounds, shown in Table 2.3.-1. were mainly quantified using the peak area of each target compound in the FID-chromatogram and the respective FID response factor. The quantification of halogenated compounds was however based on MS total ion-chromatogram due to their insufficient response in FID. Additionally, xylenes and trimethylbenzenes were quantified using the response factor of toluene. Other identified VOCs were quantified using the peak area of the compound in the FID-chromatogram and the FID toluene response factor. The calculations of the response factors were done by using the following formula:

$$Rf_s = c_s/A_s \qquad (3.4.1)$$

where: Rf_s = response factor of standard compound S
c_s = concentration of standard compound S
A_s = are of standard compound S taken from FID chromatogram

For each seven *EXPOLIS* VOC samples analysed, one standard tube containing known amounts of pure target compound standard solutions was analysed. The used response factor for each compound, defined as an average of four parallel response factors, was re-determined for each delivery batch. The total volatile organic compounds (TVOC) was determined by converting the total area of FID chromatogram, from hexane to hexadecane, into equivalents of toluene.

The sample concentration was then calculated using 3.4.2. The detection limit of the used system was 1 µg/m;.

$$c_A = m_A/V \qquad (3.4.2)$$

where: c_A = concentration of compound A in the sample as respective equivalent (µg/m^3)
m_A = mass of compound A in the sample converted into respective equivalent (µg/sample)
V = sample volume (m^3)

Total volatile organic compounds, TVOC-value for each sample was determined as FID-response in toluene equivalents within the range from hexane to hexadecane which is the boiling point range considered to be covered by Tenax TA-sorbent. TVOC concentration was then calculated according to the formula 3.4.2. Using this detection sensitivity, typically 40 to 70 VOCs were identified from air.

Data Transfer

The VOC results analysed at VTT were then sent back to *EXPOLIS* sampling Centre as an Excel-file to be included into local *EXPOLIS* Database. Each file contained the results (ng/sample) of one analysis set, i.e. tube batch, which was sent by respective *EXPOLIS* sampling Centre..

Active Charcoal Tubes

For preparation, the active charcoal tubes were rinsed with 0.5 mL carbon disulfide (CS_2) and dried under a pure nitrogen gas flow (10 min). They were tightly sealed with caps and stored in a closed glass container. Prior to use, the sampling tubes were reconditioned by rinsing them twice with 0.5 mL CS_2 and by drying them under a pure nitrogen gas flow (10 min). The clean and sealed active charcoal tubes were stored in a closed container for less than one week before sampling.

After sampling, the exposed tubes were stored at 4°C (0-3 days). An internal standard solution (6µL) was added to both sections of charcoal. VOC's were desorbed from the charcoal sampling tubes with 300µL (1st step: 250 and 2nd step: 50 µL) carbondisulfide for each section into one vial with 2 mL volume. The vials were closed and stored at 4°C until analysis (1-7 days).

The gas chromatograph was calibrated using 8 different calibration solutions (diluted from stock solutions), containing known amounts of pure standard solutions and internal standard. Stock solutions could be stored in the freezer for at least 9 months. The calibration solutions were counterchecked every 3 months by an another laboratory (Umweltschutzlabor Liestal, Switzerland).

For analysis, the samples were loaded sequentially into an autosampler with the first tube recommended to be a clean one. The second one was the first calibration standard. The 8 standard solutions were run for each 10-20 samples. With the autosampler, 1 µL of the eluate was injected into the GC equipped with a tandem capillary column (injector side: HP5; detector side: Innowax). The tandem column was connected to MSD by a 0.5 m transfer line (deactivated fused silica 0.2 mm i.d.).

The VOC's were identified from the MSD ion chromatogram by reference spectra and retention time. To accept the result, the quality of the match should generally exceed 500 (ITD fit).The respective peaks were quantified by peak area of the specified mass of the compound, using the calibration curve based on the 8 calibration solutions. Based on a 30 liter sampling volume, the lower limit of identification and quantification from ITD was 1-5µg/m^3.

Calculation from the analysed mass to air concentration was computed automatically in the *EXPOLIS* database. The VOC sampling flow rate was calculated as the average of sampling flow rates measured before and after sampling. The sampled air volume was calculated by multiplying the VOC sampling flow rate with total sampling time. Volumes were normalized to NTP and the VOC concentration was calculated by dividing the mass (ng) of a specific compound analyzed with GC-MSD/FID (Tenax TA) and GC-MSD (active charcoal) by the normalized sampling volume.

3.4.2. Quality Assurance/Quality Control

SOPs

Identical Standard Operating Procedures (SOPs) were used in each *EXPOLIS* Centre to assure the quality and comparability of the data collected in different Centres. The SOPs were developed for all laboratory, field and subject procedures used in the study.

The SOPs developed for the VOC sampling and analysis in *EXPOLIS* are:

- **VOC sampling (Expolis/VTT-F-4.0)**, defines the VOC sampling setup, flow calibration and other preparations in the laboratory before and after the sampling.
- **VOC sample analysis (Expolis/VTT-L-1.0)**, defines the VOC sampling tube preparation and tube analysis procedures in the analytical laboratory.
- **VOC sampling and analysis QA (Expolis/VTT-L-4.0)**, defines the quality assurance procedures for the VOC sampling and analysis.

Intercalibration

A 1st intercalibration exercise was performed in August 1996. A VOC mixture containing 11 compounds (n-hexane, toluene, tetrachloroethene, n-octane, 2-butoxyethanol, _-pinene, 1,2,4-trimethylbenzene, 1,4-dichlorobenzene, n-decane, 2-ethyl-1-hexanol, n-dodecane), was prepared in the 30 m^3 environmental test chamber, named INDOORTRON, at the EC-JRC Environment Institute (JRC-EI), Ispra, Italy. The amount of each compound, be it liquid or solid, was weighed and the resulting liquid mixture injected into the supply air stream of the chamber. This injection was realised through a laboratory-built device consisting of a reservoir, a liquid chromatography pump, a fused silica capillary and a heated sintered stainless steel disk, equipped with a temperature sensor. The short temperature negative peaks caused by the falling drops of the mixture were continuously recorded: the frequency and the mean weight of the drops enabled to accurately determine the delivery rate of the solution. The air flow rate through the chamber was controlled by frequent injections of SF_6 tracer and found satisfactorily constant; also the efficient mixing of the vapours with the chamber air was checked by sampling this tracer at different positions: no increase of the overall variance was observed in the tracer concentrations when measuring at the different positions, compared with the measurement at a single position.

To obtain the concentrations, the composition of the mixture must be known. In addition to the knowledge given by the original weights of the different compounds, this information was obtained analysing the mixture contained in the reservoir of the injection device at the beginning and at the end of the injection period. A slight change in the composition of the mixture over time due to evaporation was observed: apart from n-hexane (which was excluded from the comparison), the changes were within ∀5% from the original composition. Concentrations between 50 and 150 $\mu g/m^3$ (200 for

tetrachloroethene) were produced in the chamber.

VTT used Tenax TA samplers for thermal desorption supplied by the JRC-EI, Carbotech used active charcoal samplers of their own. Three Tenax TA samplers were loaded with one litre of chamber air each, and three active charcoal samplers with 8-9 litres each. In addition, one blank (unloaded) sampler was mailed to each laboratory. The laboratories had no knowledge about the mixture, except that the concentrations were greater than 50 $\mu g/m^3$.

On the basis of the results of the 1st comparison, a 2nd intercalibration exercise was realised in April 1997, with much smaller VOC amounts to simulate real samples. In view of the difficulties and uncertainties in preparing, in the INDOORTRON test chamber, a gaseous mixture at the concentration level desired ($\approx 10\ \mu g/m^3$), it was decided to use a liquid mixture and to deposit 1 µl thereof onto each sampler. In order to have a better understanding of the results, a second solution was used, which produced masses on the samplers similar to those of the 1st comparison, i.e. roughly 10 times higher than the first solution. There were 8 compounds in this 2nd exercise (cyclohexane, toluene, hexanal, tetrachloroethene, 2-ethyl-1-hexanol, limonene, undecane, naphtalene) and they were partially different from those used in the 1st exercise. Compounds were selected with the criterion of having at least one compound for each of the six classes of *EXPOLIS* target compounds. Because of the different analytical systems (VTT as well as JRC-EI using thermal desorption with whole sample injection, Carbotech using solvent desorption with injection of 2 µl out of 300 µl) also the solutions needed to be different: Carbotech samplers were loaded with amounts 50 - 100 times higher than JRC-EI and VTT samplers. Methanol was used as solvent and it was volatilized from the samplers with 1 l of high purity He.

Each laboratory was requested to mail 6 samplers to JRC-EI (2 for each solution plus two as blanks); after loading, they were returned to the laboratory: altogether it took less than one week for the samplers from leaving to returning to the respective laboratory.

Field Blanks and Duplicates

About 10% of all PEM and MEM VOC field samples were accompanied with field blanks. These were complete VOC sampling assemblies which went through all the procedures with the actual samples, except that they were not connected to pump for actual sampling.

The determination of VOC sampling precision based on field duplicate measurements. The duplicate PEM samplers were carried by the researchers. The duplicate MEM samplers were taken to the field along the normal measurement. The number of field duplicates were about 10% of all VOC samples. Field blank and duplicate VOC samples were included in the measurements throughout the whole field work period.

3.4.3. Results

VOC Sample Analysis

The overall results of the VOC analyses for Athens, Basle, Helsinki, Milan and Prague are presented in Tables 5.5.2 A, B, H, M and P.

In Table 3.4.-1., theoretically calculated actively sampled and passively diffused TVOC-masses for typical *EXPOLIS* MEM (Tenax TA) sampling arrangement without and with diffusion barrier are shown. The passive mass transfer through a layer of gas is calculated using Fick's first diffusion law. The following assumptions are used:

- TVOC-concentration = 500 $\mu g/m^3$,
- Active VOC sampling flow = 2 mL/min,
- Active sampling time = 2*8 h = 16 h,
- Time between measurement set-up and download (= passive sampling time, active sampling time excluded) = 40 h, and
- Diffusion coefficient for TVOC = 0.07 cm^2/s.

As shown, theoretically the diffusion barrier system limits passive diffusion to 0.25% of the situation when the diffusion barrier system is not used. Also when the diffusion barrier system is used the amount of passively diffused VOC mass when compared to actively sampled mass is meaningless (0.05%).

For the 30 compounds studied, the percentage of samples above the sample detection limit (LOD) for Helsinki Tenax TA and Basle active charcoal PEM and MEM samples are given in Table 3.4.-2. For Helsinki samples 17 compounds were found in 50-100 % of personal samples. For MEM-in, MEM-out and MEM-work samples the number of compounds found in 50-100 % of samples were 19, 9, and 15, respectively. Compounds not found in any sample were for personal and MEM-work measurements 1-octanol and 1,1,2-trichloroethane and for MEM-out measurements 2-buthoxyethanol and 1,1,2-trichloroethane. For MEM-in samples every target compound was found at least one sample.

For Basle personal, MEM-in, MEM-out, and MEM-work samples the number of compounds found in 50-100 % of samples were 15, 16, 2 and 8 (of 24 compounds, 6 targets cannot be determined with active charcoal method), respectively. Compounds not found in any sample were for personal and MEM-work measurements 1,1,2-trichloroethane, for MEM-in measurements styrene and 1,1,2-trichloroethane and for MEM-out measurements nonane, undecane, styrene, propylbenzene, benzaldehyde, and 1,1,2-trichloroethane.

QA/QC

Intercalibration

Hexane is excluded from the results of the 1st intercalibration exercise because of the too high changes in the liquid mixture during the experiment.

The VTT results for five compounds were within 10% from the expected concentrations, whereas the other five deviated more; all on the low side. Particularly large is the discrepancy for butoxyethanol, for which however also the JRC-EI had a difference of -9%, compared with the expected concentration. The results by Carbotech were within 10% of chamber concentrations for three compounds; three other concentrations were not reported and for the remaining four compounds, deviations were up to 30%. The standard deviations associated with the triplicate determinations in general were high.

There are two discrepancies to be noticed before considering the individual results of the 2nd intercalibration exercise. The first one concerns undecane, for which the mass measured is about 1/3 of the mass nominally deposited. The cause for this discrepancy was discovered after the analyses at the JRC-EI: the composition of the liquid contained in the bottle was different from the composition declared on the label. A 2nd bottle with identical lot number turned out to give the same inconvenience. A possible explanation is that, due to an error, the bottles of this lot have not been filled with pure n-undecane, but with the mixture of "impurities" from GC-MS analysis as alkanes, cycloalkanes and alkenes. The second discrepancy concerns cyclohexane: the mass of this compound found on Tenax TA samplers is much lower than the mass deposited because a large fraction was volatilized along with methanol. This, in turn, is due to the fact that 1 l of He was used for the removal of the solvent; this volume was adopted considering that up to 3 l of air are currently sampled within the project.

The VTT results were very close to those obtained by JRC-EI. For the more concentrated solution, a deviation > 10%, when compared to loaded amounts, occured only in the case of hexanal (- 13%); for the more diluted solution, there was only one compound with a deviation largely exceeding 10% (tetrachloroethene +24%) and three compounds with borderline deviations (toluene + 11%, hexanal - 10.4% and 2 -ethyl-1-hexanol + 10.6%). The standard deviations associated with the duplicate measurements, excluding cyclohexane, were <4% for the more concentrated solution and <6% for the more diluted solution.

In the results by Carbotech, 2-ethyl-1-hexanol was not detected at all, even in the more concentrated solution; naphtalene was heavily underestimated when compared to loaded amounts (-54% and -89%, respectively for more concentrated and more diluted solution) and the same happened for hexanal in the more diluted solution (-95%). However, there was a general underestimate of 15-35% also for the other compounds. Concerning the standard deviations from the duplicate analyses, they were extraordinarily high for the two polar compounds determined (hexanal and naphtalene), but they exceed 10 % also for cyclohexane and limonene in more concentrated solution (19% and 24% respectively).

Field Blanks and Duplicates

The median background and 95th percentile levels of VOC field blanks (71 for Tenax, 40 for Carbotrap) are shown in Table 3.4.-3. The median background levels of target compounds, determined from analysis of field blank sampling tubes, were for Tenax TA samples less than the sample detection limit for all 30 target VOCəs. At a nominal 2-4 L of air collected per cartridge, this corresponded roughly to concentration of less than 1 $\mu g/m^3$. Also only for three target compounds, toluene, benzaldehyde and octanal, the 95th percentile level of Tenax TA field blanks exceeded the sample detection limit.

With active charcoal samples the background levels of target compounds were in nanograms higher than with Tenax TA samples, but when the larger sampled air volume with charcoal tubes (30-50 L) were observed, the median background contamination found in field blanks was approximately the same as with Tenax TA samples (less than 1 $\mu g/m^3$).

The agreement between duplicate sample pairs is summarized in Table 3.4.-4. for target compounds found above detection limits in both samples. The per cent relative standard deviation (% RSD) for those pairs is calculated and the median and 75th percentile reported. The number of Tenax TA duplicate pairs available was 15 for personal samples and 46 for microenvironmental samples. The median RSD ranged for personal samples from 1.7 (3-carene) to 21.4 % (2-ethylhexanol) and for microenvironmental samples from 1.9 (2-buthoxyethanol to 38.4 % (phenol). In personal exposure measurements for next target compounds no duplicate pairs with both samples above sample detection limit were found: phenol, 1-octanol, trichloroethene, 1,1,2-trichloroethane, 1-methyl-2-pyrrolidinone. For 1,1,2-trichloroethane and 1-methyl-2-pyrrolidinone this was true also in microenvironmental measurements.

3.4.4. Discussion and Conclusions

VOC Sample Analysis

The intercalibration results show that the method of long duration (...48 h) and ultra low flow rate (0.5 ... 2 mL/min) active sampling in Tenax TA, thermal desporption, GC separation and tandem analysis by MSD (qualification) and FID (quantification) is applicable for the determination of the selected 30 target VOCs in outdoor and indoor air with a possible exception of the very volatile hexane. The data Tables 5.5.2 A, B, H, M and P demonstrate the sensitivity (about 1 $\mu g/m^3$ for most compounds) of the method and its dynamic range (1...1.000 $\mu g/m^3$). The results by Carbotech AG confirm that the method is inadequate for some compounds. As expected, 10 of the 30 *EXPOLIS* target compounds could only be determined qualitatively and for 5 compounds the method is not suitable at all. 16 compounds could be quantified. (see Table 3.4.-2).

For nearly all target compounds the percentage of samples above the sample detection limit were higher for Helsinki samples than for Basle samples when outdoor samples were compared. This doesn't mean that the outdoor air in Helsinki is more polluted when the concentrations of VOC's are compared, but it means that the Tenax TA sampling with thermal desorption is more sensitive sampling method with a lower sample detection limit than the active charcoal sampling with liquid desorption for the most of target compounds. Only with few compounds as 1-methyl-2-pyrrolidinone the charcoal sampling with liquid desorption seems to be more sensitive sampling method.

The problem of the flow rate increase with increasing particulate filter loading could have been prevented by drawing the air before and not after the filter. However, assuming a linear flow rate increase, this did not create a systematic error of the sample volume which was calculated with the mean flow measured at the beginning (unloaded "day" filtre) and at the end (loaded "nigh" filtre) of the sampling period. The higher VOC-flow rate changes in the MEM sampling reflect the about four times higher $PM_{2.5}$ flow rate in MEM sampling compared to PEM sampling.

QA/QC

Intercalibration

The results of the 1st intercalibration exercise were unsatisfactory. With butoxyethanol, in the tests carried out successively at the JRC-EI with two samplers in series, it has been noted that butoxyethanol may present a certain break-through, even sampling 1 liter; the amount is not reproducibile, but the fraction found on the 2nd sampler never exceeded 6% of the amount found on the 1st one. In fact, the data show a behaviour different from typical chromatographic break-through and the phenomenon disappears with dry air; this seems to indicate an entrainment of butoxyethanol (and may also be of other hydrophilic compound) by water.

According to the 2nd intercalibration exercise, in the case of Tenax TA sampling of cyclohexane, results with the observed loss points to the need of carefully verifying for which of the target compounds the break-through volume is smaller than 3 litres, i.e., which compounds are not collected quantitatively when sampling 3 liters of air. This inconvenience will not happen with activated charcoal samplers.

When undecane and cyclohexane are excluded, the VTT results of the 2nd intercalibration, considering the masses analyzed (_ 10 ng), may be considered satisfactory. The standard deviations associated

with the duplicate measurements, excluding cyclohexane, are < 4% for the more concentrated solution and <6% for the more diluted solution.

The results by Carbotech show that the method is inadequate for polar compounds. In the case of hexanal and naphtalene, it should be noted that concerned compounds are only moderately polar compounds.

Field Blanks and Duplicates

Background levels of target compounds, determined from analysis of field blank sampling tubes, were for Tenax TA sampling tubes low. The median background levels for all 30 target VOCэs were smaller than the limit of detection (LOD). The duplicate results show that the method produces repeatable results for the target compounds (mostly within 10%, Table 3.4.-4). The stainless steel capillary diffusion barrier and careful sampling tube treatments effectively limited tube contamination to LOD for almost every sample and compound (Table 3.4.-4.).

In the New Jersey TEAM-study (field measurements were done 1981), background levels of Tenax GC field blanks were usually less than 20 ng/cartridge for all 20 target compounds with the exception of benzene (97 ± 64 ng/cartridge) and 1,1,1-trichloroethane (33 ± 21 ng/cartridge) (Wallace *et al.*, 1986). At a nominal 20 L of air collected per cartridge, this corresponded to concentration of less than 1 $\mu g/m^3$. In the Los Angeles TEAM-study (field measurements were done 1987), the measured amounts of blank Tenax GC cartridges were in the range of 0-10 ng (the equivalent of 0.0-0.5$\mu g/m^3$) for all 25 target compounds except benzene (5-33 ng), chloroform (7-42 ng), 1,1,1-trichloroethane (1-36 ng) and limonene (0-27 ng) (Wallace *et al.*, 1991). The personal and microenvironmental sampling periods were in TEAM-study 12-hour/sample and the sampling was carried out continuously without any breaks.

In the study by Wallace *et al.* (1989), the influence of personal activities on exposure to VOC's were studied. Personal and microenvironmental evening, night and day VOC samples were collected with Tenax GC. Blank cartridges showed contamination levels of 0-2 ng/cartridge for all 17 target compounds except benzene (8 ng/cartridge) and chloroform (6 ng/cartridge). At the nominal sampling volume of 7 liters, the latter two correspond to concentration of 1 $\mu g/m^3$.

De Bortoli *et al.* (1992) analyzed 23 freshly cleaned Tenax TA tubes and found that benzene and toluene contributed most to the background emission of sorbent. The 50th and 90th percentiles of background emissions for 100 mg of Tenax TA sorbent were for benzene 5.6 and 16 ng and for toluene 1.9 and 5.0 ng, respectively.

When compared to above mentioned studies the results of *EXPOLIS* blank samples can be considered as good as the results achieved in earlier remarkable VOC exposure studies where sampling were carried out with Tenax GC sorbent.

In the New Jersey TEAM-study, the median coefficient of variance (=RSD) of field Tenax GC duplicates ranged from 20 to 40 % in most cases (Wallace *et al.*, 1986). The highest median CV's were found with benzene. These were for personal samples 36 and for outdoor samples 47 %. In the California TEAM-study (field measurements were done 1984) (Hartwell *et al.*, 1987), median CV of Tenax GC samples ranged from 7 to 23 % for personal and from 10 to 34 % for outdoor samples. The highest median CV for personal samples was found with 1,4-dioxane and for outdoor samples with chloroform. In the 1987 TEAM-study in Los Angeles (Wallace *et al.*, 1991, Hartwell *et al.*, 1992) the median RSD of Tenax GC duplicates ranged from 10 to 20 % in most cases. Overall, in this study the number of duplicate pairs with measurable data was quite small.

In the study by Wallace *et al.* (1989), the precision of Tenax GC duplicate air and breath samples ranged from median RSD of 5 to 17 %. In the VOC exposure study while commuting (Chan *et al.*, 1993) the relative mean deviation of duplicate samples ranged from 4.8 to 9.2 % for nine target VOC's. In the other study by Chan *et al.* (1994) the relative mean deviation of Tenax GC duplicates was also within 10 % for all target compounds. In the study by Lawryk and Weisel (1996) the VOC samples

were collected by suspending adsorbent tubes containing either Tenax or a layered adsorbent (Tenax GC, Carboxen 569 and Carbosieve SIII). The mean RSD of duplicates were nearly all target compounds less than 20 %, with the exceptions of the two light alkanes, 3-methylpentane (23 %) and n-hexane (48 %). In the study by Baek *et al.* (1997) the VOC samples were collected with Carpotrap (60/80). In this study the precision of the sampling of 8 target VOC's ranged from 2.5 % (benzene) to 5.0 % (ethylbenzene). Precision was expressed as a RSD of at least ten side-by-side analyses of a standard sample.

When compared to above mentioned studies, the results of *EXPOLIS* duplicate samples can be considered as good as the results achieved in these studies.

The methods used in the *EXPOLIS* to determine human exposure to VOCs, and the quality assurance to control measurement quality were developed especially for this project. As shown above, the methods have proven to be reliable and applicable for sampling exposure levels at low VOC concentration levels. In general the results seem to show that the consistent quality assurance work done at every stage of the VOC sampling and analysis will be paid off.

3.5. CO Monitoring

3.5.1. Methods

In *EXPOLIS* study carbon monoxide (CO) was selected to represent exposure to traffic exhaust and indoor combustion sources. CO and temperature were measured using Langan Model T15 High resolution personal exposure monitor. CO PEM consisted of the electrochemical sensor with electronics, DataBear data logger, and internal and external temperature sensors.

The electrochemical CO sensor was manufactured by City Technology Ltd (Portsmouth, England). The air flow to the sensor was passive (no pump). The sensor detected the chemical reaction of CO to CO_2. This reaction generated an electrical current proportional to the CO concentration. In the measurements of ambient level CO other gases are not normally present at levels which could cause response for the sensor. For example 3.3 ppm of NO_2 will result 0.5 ppm increase in measured CO concentration and 25.0 ppm SO_2 will result 0.5 ppm increase in measured CO concentration (Langan, 1992). Interference of other air pollutants was hardly a problem in the *EXPOLIS* CO measurements. For example in Helsinki one hour average levels in ambient air for NO_2 were 0.05-0.17 ppm and for SO_2 0.01-0.05 ppm in year 1997 (Aarnio *et al.*, 1998).

In *EXPOLIS* four channels of DataBear were used:

- Cannel 1 measured CO with measurement range 0-128 ppm and 0.5 ppm resolution
- Channel 2 measured temperature adjacent to CO sensor in degrees Fahrenheit (_F)
- Channel 3 measured CO with measurement range 0-12.75 ppm and 0.05 ppm resolution
- Channel 4 measured temperature of external sensor in degrees Fahrenheit (_F).

Temperature corrected CO concentration was needed because electrochemical sensor and electronics are temperature dependent. The external temperature sensor was used to validate Time Location Activity Diary data and calculate the sampled volume of the Buck I.H pump in $PM_{2.5}$ PEM sampling. Checking DataBear settings and data downloading were done with *EXPOLIS* specific program Langan.EXE. After measurement it was drawn a chart to check that measurement was all right. Batteries were checked also by Langan.EXE program.

Zeroing and calibration for each Langan was done at least once a week with zero gas and CO

calibration gas. The observed zero value had to be 0-1.5 ppm and ± 0.5 ppm of the previous zero value. The observed calibration value had to be ± 25% of the previous calibration value. CO PEM external temperature sensor was checked at least once a month against thermometer and it was not allowed to have larger difference than ±2.5 _C.

In *EXPOLIS* the subjects carried the monitor with them in the PEM sampler along with the other personal measuring devices for two consecutive weekdays. CO PEM measured continuously and it was not changed during the measurement days like filters in personal $PM_{2.5}$ sampling. CO concentrations and temperature values were collected by 1 minute interval. Separate microenvironmental CO measurements were not done for the subjects of *EXPOLIS*. Additional microenvironmental measurements of CO were measured in traffic, restaurants, supermarkets etc by the researchers.

CO concentration is calculated as follows:

$$\chi_c = \text{calibration factor} * (\chi - \chi_{ero}) \qquad (3.5.1.)$$

where: calibration factor $=\chi_{bottle}/\chi_{calibration}$
χ_{bottle} given CO concentration of the bottle (ppm)
$\chi_{calibration}$ = measured CO concentration of the monitor in calibration (ppm)
χ_i= measured CO concentration in the field (ppm)
χ_{zero} = measured CO concentration in zeroing (ppm)

Conversion factor: 1 ppm CO = 1.164 mg/m^3 @ 20 oC
These calculations and calibration corrections are done with the computer program but in this report temperature corrected values are not used.

QA/QC

Duplicates and comparison to NDIR (non dispersive infrared analysis, the standard reference method for CO in ambient air)

The CO duplicate measurements were done same way as PEM $PM_{2.5}$ duplicates. The PEM samplers including CO monitors were either carried by the researchers, placed in one location or included in separate microenvironmental measurements done for example in transport vehicles nominally for 24 hours. The target number of duplicated measurements was 5% of all the measurements during the field period in all Centres.

Langan monitor was compared to the fixed station CO monitor (NDIR). It was done at least once during field period and at least in two Centres.

Intercomparison of the instruments from different centres

CO monitors from each centre were tested side by side in Helsinki in connection with the second *EXPOLIS* Workshop. Each Centre brought one of their Langan monitors that they were using in their measurements. Each Centre performed the zeroing, calibration and data downloading by themselves. CO sample interval was one minute during 42 hours measurement period and Langan monitors were located side by side. There were some technical problems with the Langan monitor from Milan first and this is why it was tested again later against one of the Langan monitors in Helsinki.

3.5.2. Results

Pilot

The CO measurements worked out very well. Data channel 3 (more precisely CO channel (0.05 ppm resolution)) was very important because in most cases the concentrations remained low. As a consequence of occasional data losses the download procedure was changed so that data was downloaded when Langan was still inside the PEM case. The additional external temperature sensor was found to be more useful (faster responding) for Time Location Activity Diary data validation than the standard internal temperature sensor. In the duplicate measurements it turned out that it is important that the clocks of the compared CO monitors are exactly in the same time.

QA/QC

SOPs

Based on the pilot experiences the procedures were finalized and written into SOPs. The following SOPs were most relevant for CO monitoring:

MEM sampler positioning and PEM sampler carrying SOP describes how the home indoor, home outdoor and workplace MEM samplers should be located in various personal and microenvironmental settings and how the subject should keep the PEM sampler in different circumstances, and carry it when moving.

CO monitoring SOP describes the procedures for calibration, zeroing, programming, field application, data loading and quality control of the Langan T15 CO personal monitor for collecting continuous personal CO and temperature exposure data.

CO monitoring QA SOP describes the general procedures for fixed monitoring field comparison and duplicated measurements of the Langan T15 CO personal monitor in *EXPOLIS* personal CO measurements.

Intercomparison Test of Instruments from Different Centres

The intercomparison test of the CO monitors from Athens, Basel, Helsinki and Prague are presented in Figure 3.5.-1.

The average CO concentration for all Langan monitors was 1.5 ppm (range 1.4 B 1.6 ppm, standard deviation (sd) 1.1 ppm) (see Table 3.5.-1.). The average temperature was 23.7 °C and its standard deviation was 0.3 °C. There was no observable difference on the high end of the measurement (99 % of measurements were in range below 4.7-4.9 ppm) or low end (25 % of measurements were in range below 0.3-0.5 ppm).

In a later intercomparison between the CO monitors of Milan and Helsinki average CO concentration for Milan was 1.2 ppm and for Helsinki 1.4 ppm, sd for both instruments was 1.3 ppm. There were no

difference on the high end of the measurements (99% of measurements were in range below 5.5-5.9 ppm) or low end (25% of measurements were in range below 0.1-0.3 ppm) (see Table 3.5.-1). There was a slight difference in the observed temperature values. Average temperature for the Langan monitor of Milan was 23.0 °C and that of Helsinki 22.7 °C. SD for both was 0.3 °C.

Correlation between the Langan monitors of different Centres' varied between 0.997-0.998 (see Table 3.5.-2.).

In addition to these intercomparisons, 11 duplicate measurements were done in Helsinki and 10 in Basel during the field work. The agreement between the duplicates was generally excellent, repeating the intercomparison results.

3.5.3. Discussion

The Langan monitor is easy to operate, lightweight, noise free and has a large memory capacity, which make it quite suitable for personal measurements.

The CO monitoring SOPs appear to have ascertained excellent CO data comparability between the Centres.

Test of instruments from different Centres

There were only very slight differences in CO concentrations of Langan monitors during the intercomparison measurements. There are all the reasons to believe that Milan′s Langan monitor is also working at the same way than others although it was tested only against Helsinki′s Langan monitor. The little differences in the CO concentrations during the measurement periods were due to different zero level. This is why the seemingly simple task of zeroing a CO monitor has to be made very carefully. The correlation between the monitors of different Centres are all very high, and we can assume that data of CO monitors is very comparable between different countries if zeroing and calibration is done according to the same standards. Temperature data of the monitors shows also only very small differences.

Table 3.3.-1. Calculated effects of the changes in the weighing room conditions to the weighted masses caused by the buoyancy effect. In this example, the assumed baseline weighing room climate is P= 740 mmHg, R.H.= 40%, T= 20 °C, the filter density is 800 kg/m^3, PEM and MEM filter masses are 100 mg and 120 mg, respectively.

	ΔP(mmHg)	ΔR.H.(%)	ΔT(°C)
Cond. Change	+30	+20	+2
Corresponding change in balance reading, μg			
37 mm PEM filter	-5.9	+0.25	+1.1
47 mm MEM filter	-7.1	+0.32	+1.3

Table 3.3.-2. The standard deviations of the 37-mm Gelman Teflo filters and the stainless steel standard weight when weighing with a Mettler MT5 Faraday cage ('Faraday'), with a small weighing pan without deionization ('Pan') and with a weighing pan using Po-210 deionizer ('Po').

Standard deviation, μg (number of measurements)			
Weighed object	Faraday	Pan	Pan + Po
37 mm filter	5.9(50)	23(25)	1.7(25)
Standard weight	3.0(17)	0.9(25)	-

Table 3.4.-1. Theoretically Calculated Actively Sampled and Passively Diffused TVOC-masses for Typical *EXPOLIS* Tenax TA MEM Sampling Arrangement Without and With a Diffusion Barrier Used. Assumed TVOC-Concentration is 500 $\mu g/m^3$.

	Actively Sampled TVOC-mass (ng)	Passively Diffused TVOC-mass (ng)	Passively Diffused / Actively Sampled (%)
Without Diffusion Barrier Tube: L = 50 mm, i.d. = 5 mm	960.0	197.9	20.61
With Diffusion Barrier Tube: L = 200 mm, i.d. = 0.5 mm	960.0	0.5	0.05

Table 3.4.-2. Percentage of Samples Above the Sample Detection Limit (LOD)

Compound	Tenax Ta / Helsinki				Active Charcoal / Basle			
	% of Samples				% of Samples			
	PEM	MEM-in	MEM-out	MEM-work	PEM	MEM-in	MEM-out	MEM-work
Hexane	21.8	10.5	37.5	32.5	42.0	34.0	27.7	30.8
Nonane	74.6	82.1	56.0	55.6	62.0	55.3	0.0	38.5
Decane	76.7	79.5	41.1	57.0	96.0	89.4	27.7	66.7
Cyclohexane	23.3	19.5	4.2	15.9	-[a]	-[a]	-[a]	-[a]
Undecane	91.7	91.6	32.7	62.3	88.0	80.9	0.0	51.3
Benzene	86.5	71.1	83.9	76.8	70.0	59.6	27.7	43.6
Toluene	99.5	99.5	95.8	99.3	100.0	100.0	78.7	94.9
Ethylbenzene	96.9	93.2	62.5	80.1	80.0	68.1	25.5	33.3
m(&p)-Xylene	99.5	98.4	86.9	96.7	100.0	97.9	78.7	87.2
o-Xylene	92.8	92.6	67.3	77.5	94.0	78.7	40.4	53.8
Styrene	39.4	50.0	6.6	19.2	20.0	0.0	0.0	12.8
Naphtalene	9.9	24.2	1.8	5.3	-[a]	-[a]	-[a]	-[a]
Propylbenzene	42.0	39.0	11.3	19.2	26.0	19.1	0.0	12.8
Trimethyl-benzenes	94.3	91.6	63.1	80.1	84.0	66.0	23.4	48.7
2-Methyl-1-propanol	45.1	69.5	2.4	17.9	-[a]	-[a]	-[a]	-[a]
1-Butanol	86.5	92.1	6.6	45.0	-[a]	-[a]	-[a]	-[a]
2-Ethylhexanol	59.1	62.6	10.1	50.3	70.0	51.1	4.3	43.6
Phenol	11.4	14.7	7.1	11.3	-[a]	-[a]	-[a]	-[a]
1-Octanol	0.0	4.7	0.6	0.0	-[a]	-[a]	-[a]	-[a]
2-Buthoxyethanol	18.1	24.2	0.0	9.3	68.0	59.6	2.1	59.0
Hexanal	96.4	96.3	23.2	62.9	100.0	97.9	23.4	89.7
Benzaldehyde	88.6	94.2	81.6	84.1	48.0	51.1	0.0	30.8
Octanal	83.4	87.4	61.3	55.6	58.0	63.8	4.3	41.0
Trichloroethene	4.7	3.2	0.6	4.0	26.0	21.3	2.1	12.8
Tetrachloroethene	10.4	9.0	0.6	6.0	22.0	17.0	6.4	17.9
1,1,2-Trichloroethane	0.0	0.5	0.0	0.0	0.0	0.0	0.0	0.0
d-Limonene	95.9	98.4	13.7	74.2	100.0	97.9	27.7	79.5
1-Methyl-2-pyrrolidinone	1.6	0.5	0.6	0.7	16.0	14.9	2.1	10.3
3-Carene	69.4	76.3	14.9	27.8	22.0	34.0	2.1	5.1
α-Pinene	96.9	99.5	33.9	65.6	72.0	76.6	6.4	41.0

[a] Cannot be determined with active charcoal method

Table 3.4.-3. Background Levels of Field Blanks

	ng/cartridge			
	Tenax Ta (N=71)		Active Charcoal (N=40)	
Compound	Median	95th Percentile	Median	95th Percentile
Hexane	<LOD	<LOD	43	112
Nonane	<LOD	<LOD	2	13
Decane	<LOD	<LOD	2	22
Cyclohexane	<LOD	<LOD	-[a]	-[a]
Undecane	<LOD	<LOD	2	19
Benzene	<LOD	<LOD	2	27
Toluene	<LOD	3.1	15	52
Ethylbenzene	<LOD	<LOD	0	28
m(&p)-Xylene	<LOD	<LOD	6	68
o-Xylene	<LOD	<LOD	0	29
Styrene	<LOD	<LOD	-[b]	-[b]
Naphtalene	<LOD	<LOD	-[a]	-[a]
Propylbenzene	<LOD	<LOD	0	8
Trimethyl-benzenes	<LOD	<LOD	2	68
2-Methyl-1-propanol	<LOD	<LOD	-[a]	-[a]
1-Butanol	<LOD	<LOD	-[a]	-[a]
2-Ethylhexanol	<LOD	<LOD	-[b]	-[b]
Phenol	<LOD	<LOD	-[a]	-[a]
1-Octanol	<LOD	<LOD	-[a]	-[a]
2-Buthoxyethanol	<LOD	<LOD	-[b]	-[b]
Hexanal	<LOD	<LOD	-[b]	-[b]
Benzaldehyde	<LOD	5.6	-[b]	-[b]
Octanal	<LOD	3.0	-[b]	-[b]
Trichloroethene	<LOD	<LOD	0	0
Tetrachloroethene	<LOD	<LOD	0	10
1,1,2-Trichloroethane	<LOD	<LOD	<LOD	<LOD
d-Limonene	<LOD	<LOD	-[b]	-[b]
1-Methyl-2-pyrrolidinone	<LOD	<LOD	-[b]	-[b]
3-Carene	<LOD	<LOD	-[b]	-[b]
α-Pinene	<LOD	<LOD	-[b]	-[b]

[a] Cannot be determined by the active charcoal method
[b] Only qualitative analysis by the active charcoal method

Table 3.4.-4. Relative Standard Deviation (%) for Duplicate Samples

Compound	Tenax TA						Active Charcoal					
	PEM (N=15)			MEM (N=46)			PEM (N=)			MEM (N=)		
	N[a]	Median (%RSD)	75 % (%RSD)	N[a]	Median (%RSD)	75% (%RSD)	N[a]	Median (%RSD)	75% (%RSD)	N[a]	Median (%RSD)	75% (%RSD)
Hexane	6	2.0	8.3	12	6.7	25.6						
Nonane	7	8.0	19.0	26	6.4	14.1						
Decane	9	8.1	18.4	26	4.7	8.0						
Cyclohexane	8	7.8	11.3	13	2.7	13.0						
Undecane	11	4.8	10.4	26	4.2	7.6						
Benzene	15	6.3	9.0	32	5.2	13.6						
Toluene	15	5.1	9.2	43	3.4	8.6						
Ethylbenzene	14	4.6	7.6	24	2.8	8.8						
m(&p)-Xylene	15	3.9	9.0	41	6.4	14.1						
o-Xylene	14	6.2	11.4	26	5.3	10.7						
Styrene	5	7.2	9.3	11	8.1	13.6						
Naphtalene	5	3.8	56.3	8	7.6	31.4						
Propylbenzene	6	5.0	7.0	12	6.4	22.1						
Trimethyl-benzenes	13	7.2	14.5	26	7.6	18.3						
2-Methyl-1-propanol	1	19.3		19	4.3	8.2						
1-Butanol	4	12.1	18.3	19	4.2	7.1						
2-Ethylhexanol	3	21.4	35.5	14	8.8	12.4						
Phenol	ND			3	38.4	56.2						
1-Octanol	ND			1	3.1							
2-Buthoxyethanol	2	18.0		3	1.9	7.9						
Hexanal	7	2.2	8.7	24	15.4	24.4						
Benzaldehyde	14	10.4	15.1	36	10.0	14.1						
Octanal	9	15.9	25.4	23	5.8	19.1						
Trichloroethene	ND			4	25.8	54.8						
Tetrachloroethene	1	7.3		4	8.4	12.8						
1,1,2-Trichloroethane	ND			ND								
d-Limonene	8	9.7	19.2	22	6.0	14.4						
1-Methyl-2-pyrrolidinone	ND			ND								
3-Carene	5	1.7	4.3	16	14.6	24.7						
α-Pinene	11	3.8	6.9	29	4.5	11.4						

[a] Number of pairs where compound found above detection limit in both samples

Table 3.5.-1. Statistical comparison of Langan CO measurements (CO and Temperature).

CO	10-12.9.1996				12-13.9.1996	
(ppm)	Basel	Helsinki	Athens	Prague	Milan	Helsinki
Average	1.6	1.5	1.4	1.5	1.2	1.4
Sd	1.1	1.1	1.1	1.1	1.3	1.3
25%	0.5	0.4	0.4	0.3	0.1	0.3
50 %	1.4	1.3	1.3	1.3	0.9	1.0
75 %	2.3	2.1	2.0	2.1	1.8	2.0
90 %	3.1	2.9	2.9	2.9	3.1	3.2
95 %	3.6	3.5	3.4	3.5	3.7	3.9
99 %	4.9	4.7	4.7	4.8	5.5	5.7
Min	0.3	0.2	0.2	0.1	0.0	0.2
Max	5.1	4.9	4.8	4.9	5.6	5.9
Temperature (°C)						
Average	23.7	23.7	23.7	23.7	23.0	22.7
Sd	0.3	0.3	0.3	0.3	0.3	0.3
Min	23.1	22.8	23.1	22.8	21.1	20.3
Max	24.2	24.2	24.2	24.2	23.1	23.1

Table **3.5.-2.** Correlations between CO concentrations of five Expolis teams during the intercomparison.

	Basel	Helsinki	Athens	Prague
Basel	1			
Helsinki	0.99828	1		
Athens	0.99845	0.99830	1	
Prague	0.99733	0.99687	0.99735	1
Milan		0.99871		

Figure 3.3.-1. Personal exposure monitor (PEM). $PM_{2.5}$ cyclone with a filter holder in left back corner, pump in the middle and battery holder in the right upper corner. Two filter holders (including filters) can be seen in the right down corner. CO monitor in the left down corner and VOC tube in the middle of the case. Black noise insulation material can be seen in the cover surface.

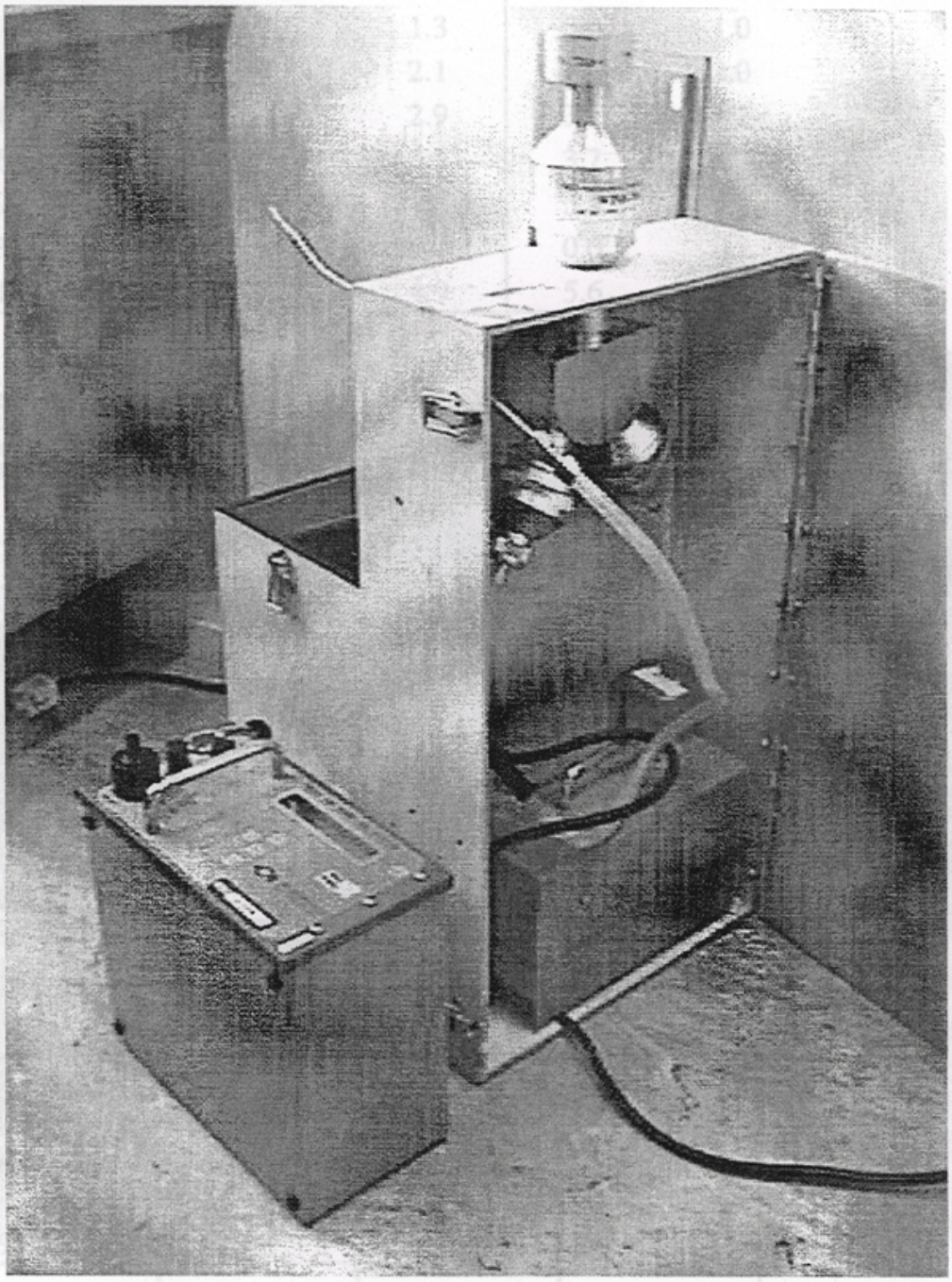

Figure 3.3.-2. Microenvironmental monitor (MEM). $PM_{2.5}$ impactor above the box, two filter holders inside the box (connected to impactor by an Y-joint), charger below the filter holders with tubing and a pump outside the box. Pump was placed inside the lower part of the box connected to hoses and the doors were closed during run periods.

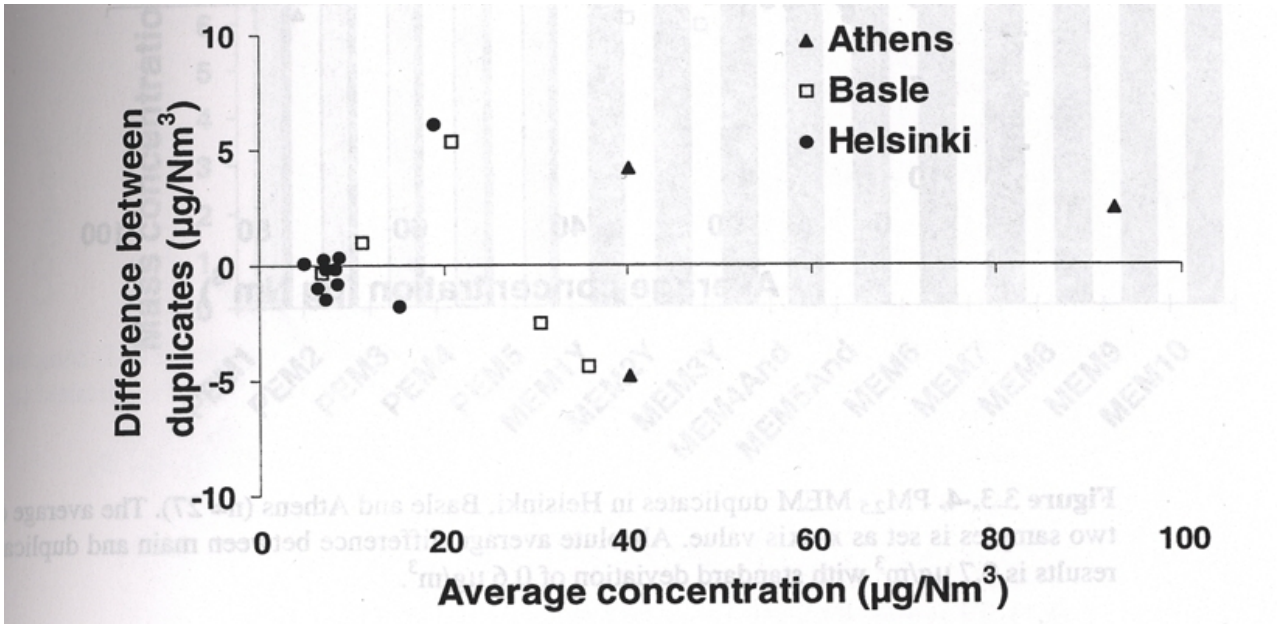

Figure 3.3.-3. $PM_{2.5}$ PEM duplicates carried by researchers in Helsinki, Basle and Athens (n= 18). The average of two samples is set as x-axis value. Absolute average difference between main and duplicate results is 2.1 µg/m^3 with standard deviation of 2.0 µg/m^3.

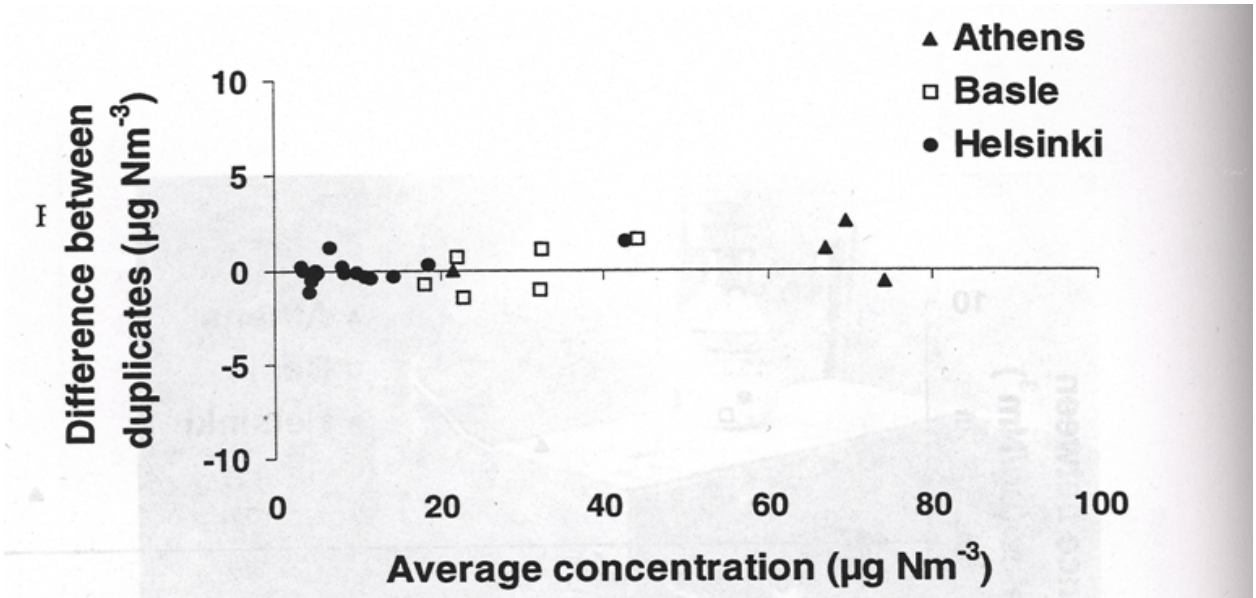

Figure 3.3.-4. $PM_{2.5}$ MEM duplicates in Helsinki, Basle and Athens (n= 27). The average of two samples is set as x-axis value. Absolute average difference between main and duplicate results is 0.7 µg/m^3 with standard deviation of 0.6 µg/m^3.

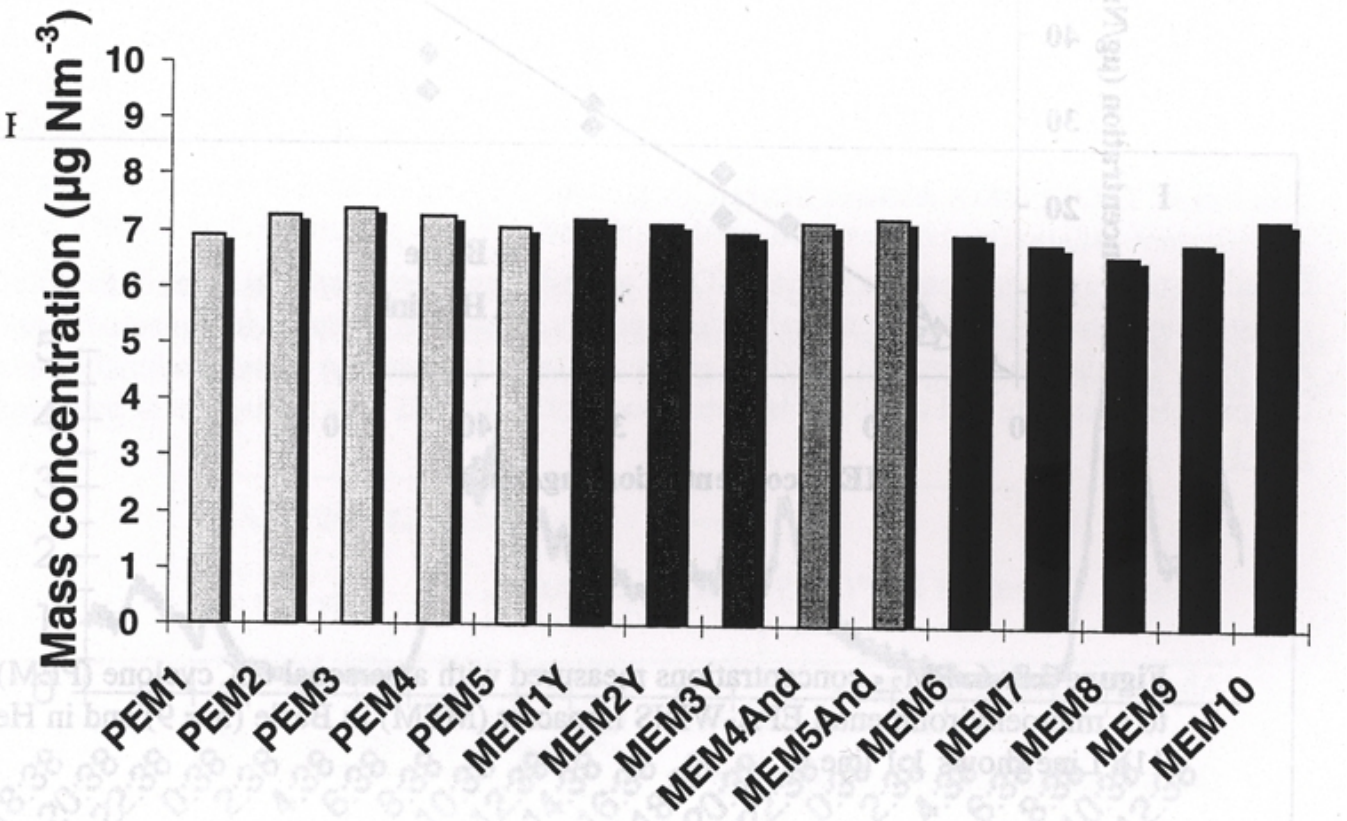

Figure 3.3.-5. PEM/MEM side by side comparison for $PM_{2.5}$. Five PEMs (PEM1-PEM5) were compared to ten MEMs, three (MEM1Y-MEM3Y) with Y-joint and two with PM_{10} preimpactor (MEM4And-MEM5And) and five indoor MEMs (MEM6-MEM10) for 45 hours.

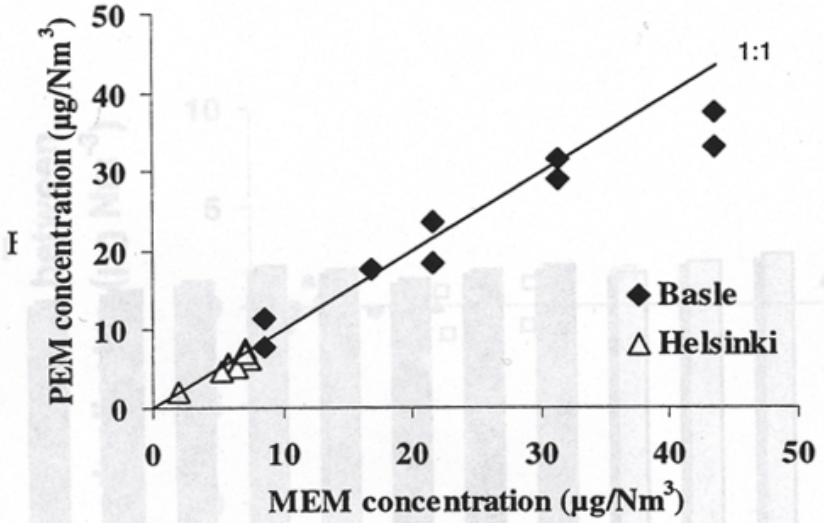

Figure 3.3.-6. $PM_{2.5}$ concentrations measured with a personal GK cyclone (PEM) compared to a microenvironmental EPA-WINS impactor (MEM) in Basle (n = 9) and in Helsinki (n = 11). Line shows 1:1 line.

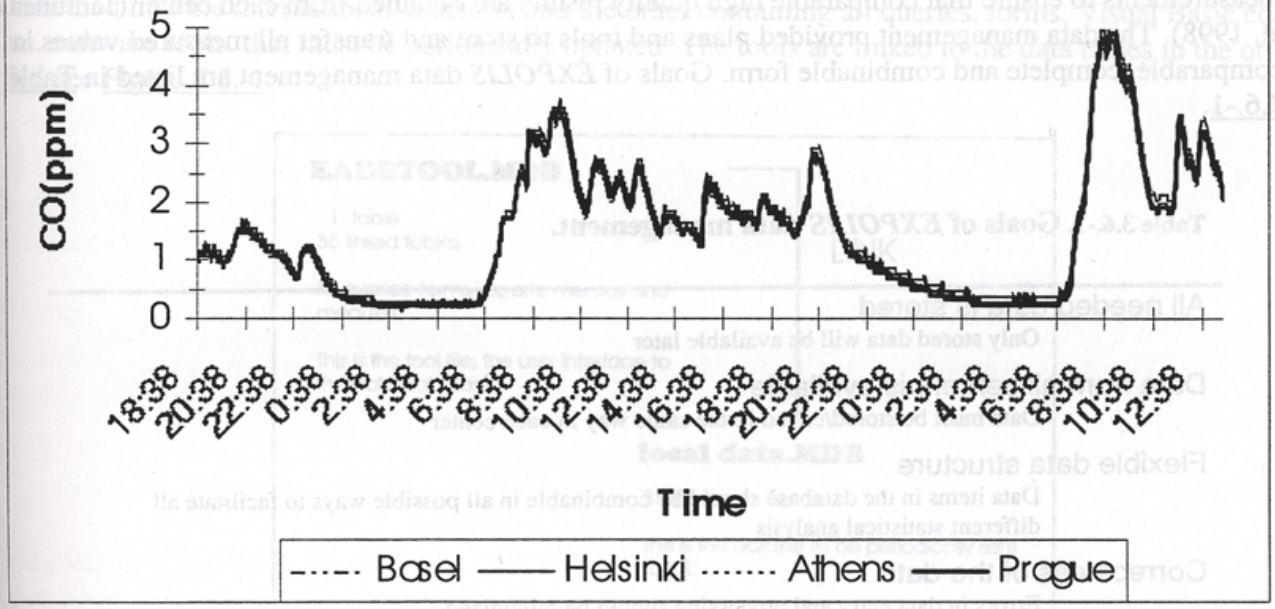

Figure 3.5.-1 CO concentrations of Basel, Helsinki, Athens and Prague CO Langan monitors during 10.-12.9.1996

3.6. Data management and the EXPOLIS Access Database (EADB)

3.6.1 EXPOLIS Data Management

Detailed standard operation procedures (SOPs) were defined for all critical tasks in the Expolis measurements to ensure that comparable high quality results are obtained from each center (Jantunen et al. 1998). The data management provided plans and tools to store and transfer all measured values in comparable, complete and combinable form. Goals of *EXPOLIS* data management are listed in Table 3.6.-1.

Table 3.6.-1. Goals of *EXPOLIS* data management.

All needed data is stored	Only stored data will be available later
Data from all centers is available	Data must be stored/coded in the same way in each center
Flexible data structure	Data items in the database should be combinable in all possible ways to facilitate all different statistical analysis
Correctness of the data	Errors in data entry and processing should be minimized
Data entry tools	Tools to enter the data into a computer must be available for each center
Privacy issues are supported	Personal information in the data must be protected and later removed

To fulfill the above listed goals, a relational database system was selected to store all measured original data items. Each data item was to be stored first on paper (serving among other purposes as 1st backup) and then in one location in the database. As each piece of data is stored only once, all later edits/corrections are completed in one step. As data is stored in the raw form, all calculations are stored in the data management system as algorithms and equations, and thus all corrections to the data, as well as to the calculation algorithms and equations are readily reflected to all results printed from the database.

3.6.2 Database Implementation and Documentation

Several commercial relational database development tools are available. Microsoft Access version 7 (MS-Office 95, Professional edition) was selected as implementation environment for the Expolis database, as it has been found flexible and powerful, as well as user friendly in many tests conducted by the computer magazines. As Access is also part of the most common office suite, MS-Office, the product itself is easy to obtain in each country, even as localized versions, and plenty of technical support is available. The program uses familiar Windows type user interface, which makes it easier to start to use a new program.

Separate tables were created in the database for each basic data type, such as air pressure, balance displays, analyzed VOC masses as well as questionnaire answers. The *EXPOLIS* Access Database (EADB) contains 36 data tables for data storage. Over 200 queries and forms were developed to facilitate the data entry, concentration calculations and other database usage.

For privacy protection purposes all information identifying a particular subject is removed, when the questionnaire, TMAD and monitoring data for the subject have been entered and quality controlled.

The basic *EXPOLIS* database was split into 2 different files, one containing all data tables, to be later combined into the international database, and the other containing all queries, forms, Visual Basic code etc. software tools, that must be periodically updated. The tools are linked to the data tables in the other file, see Figure 3.6.-1.

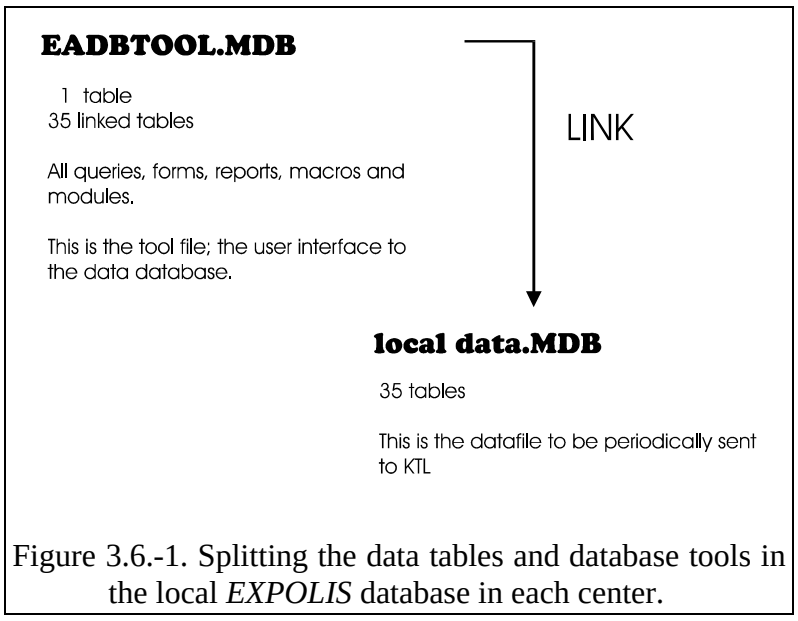

Figure 3.6.-1. Splitting the data tables and database tools in the local *EXPOLIS* database in each center.

Additional database files were created for CO 1 minute time series data, time-microenvironment-activity-diary data and ambient air quality and meteorological data (see Table 3.6.-2). The data and tool sections of these sub databases were split similarly.

Table 3.6.-2. Sections of local *EXPOLIS* database in each center.

***EXPOLIS* database sub files**		
Data file	Tool file	Contents
local.MDB (eg. HELSINKI.MDB)	EADBTOOL.MDB	questionnaires, exposures, concentrations etc.
TMAD.MDB	*TMADTOOL.MDB*	time-activity diaries,
	(to be finished)	15 minute avg. CO data
CO1min.MDB	*CO1minTOOL.MDB*	1 minute CO concentration data
	(to be finished)	
FIXED.MDB	*FIXEDTOOL.MDB*	Hourly ambient air quality data
	(to be finished)	
MET.MDB	*-"-*	Hourly meteorological data
FIXEDRUNS.MDB	*-"-*	Expolis sample sampling period averages of ambient and met data

User manuals were written for the different tasks in working with the data. The manuals are listed in Table 3.6.-3. (Hänninen et al. 1996-1998). Other minor documents were prepared as instruction sheets to describe the details of specific tasks (Hänninen 1996-1998, Hänninen and Kaarakainen 1998 and Keski-Karhu and Hänninen 1998).

Table 3.6.-3. *EXPOLIS* user manuals for the data management.

Data specification
Data entry
Concentration Query Networks PM2.5 VOC (30 compounds) NO2 *CO (to be finished)*
CO data cleaning (to be finished)
Fixed and met databases (to be finished)

3.6.3 Database Delivery and Training

The *EXPOLIS* database files were delivered and updated using mail and 3½" diskettes (files packed with PKZIP v. 2.04g), e-mail attachments and World Wide Web downloads. The final transfer of the largest data files will probably need recordable CD-ROMs or 100 MB Zip drives.

In each of the four Expolis workshops a session was dedicated for the data management and training. All practical tasks were presented and trained with researchers from each center.

3.6.4 Technical Support and Maintenance

Technical support was provided by KTL via e-mail and in some cases, with fax and telephone. The need for technical support from KTL was minimal. In some cases minor problems were caused by the localized versions of software. The English versions of all software were used in KTL to help to provide international support, but in many cases the other centers used versions with their own national language, and sometimes this made it difficult to give support in telephone or even in the workshops.

Half a dozen updates to the database tools were provided during the field stage in 1996-1997. Each update was delivered including installation instructions.

3.6.5 International Database

After the field work was finished and the data entry and cleaning completed in each center, all the local databases will be transferred to KTL (see Figure 3.6.-2.). KTL combined the data into somewhat simpler format, removing many measurement details, such as air pressure, humidity etc. data in the weighing rooms, and other variables used in the concentration calculation. Before the removal, the concentrations were calculated from the final data, and stored as static data in a separate table.

This final *EXPOLIS* database is then distributed to each *EXPOLIS* center for data queries and analyses of local as well as European *EXPOLIS* data.

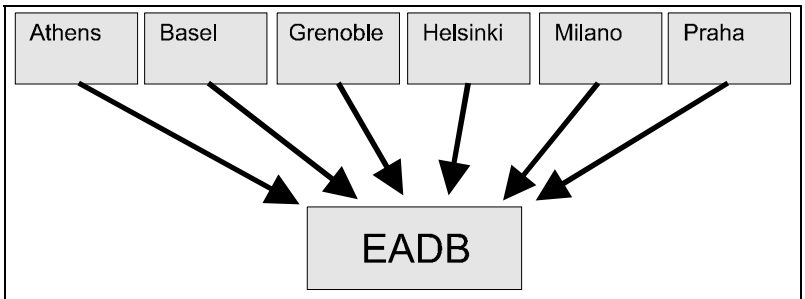

Figure 3.6.-2. Local databases from each *EXPOLIS* centers are combined into an international Expolis database.

The basic structure of the international *EXPOLIS* Access Database is shown in Figure 3.6.-3. This database contains all important *EXPOLIS* measurements from all six centers.

To maximize the utility of the complete EADB, its structure will be published in 1999, and the EADB will also be made available to research teams outside of the *EXPOLIS* team for specified analyses.

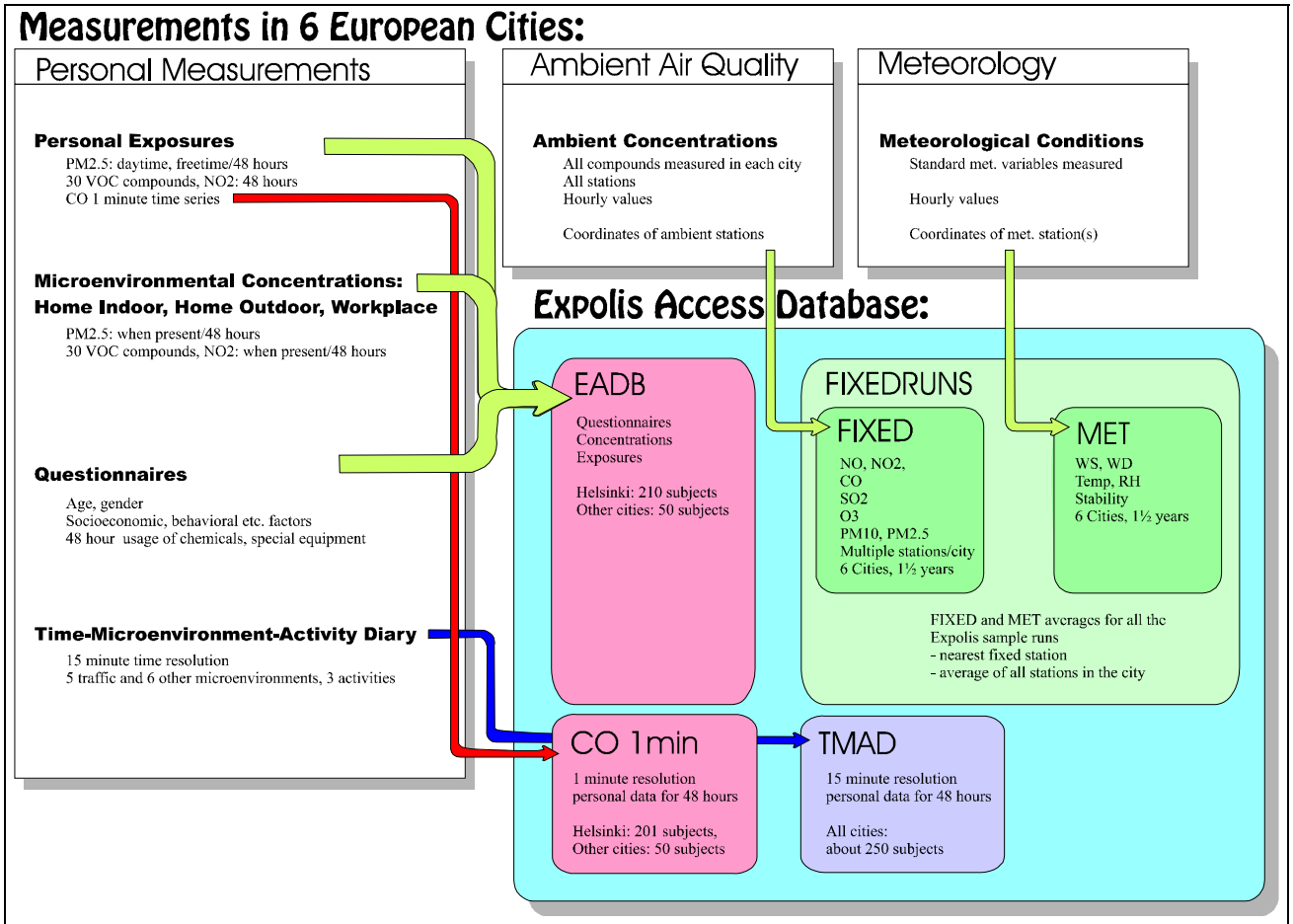

Figure 3.6.-3. Schematic structure of the international *EXPOLIS* Access Database.

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Hänninen O, Keski-Karhu J (1998): The FIXED and MET databases. Project document.

Hänninen O, Kaarakainen E (1998): Data checking and cleaning of the CO 1 minute data. Project document.

Instructions sheets:

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Hänninen O (1996): Drafting Expolis Data Management. Project document.

Hänninen O (1996): EADB Installation Instructions. Project document.

Hänninen O (1996): Joining the Access workgroup file to gain permissions to use the Expolis database. Project document.

Hänninen O (1996): LANGAN.EXE Instructions. How to download Databear data directly in Expolis format using the LANGAN.EXE download program. Project document.

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Keski-Karhu J, Hänninen O (1998): CE Distance Queries. How to calculate distances from subject's home and workplace to the fixed ambient air quality monitoring stations and to select the closest stations for each subject. Project document.

4. PRACTICAL EXPERIENCES IN THE EXPOLIS CENTRES

4.1. Athens

Field Work

Field work in Athens lasted approximately 15 months. It started in January 1997 and finished in March 1998. Overall 50 *Exposure* and 50 *Diary* subjects were completed. All 100 subjects were part of a larger (2,000 individuals) random sample of citizens from the entire Athens metropolitan area. The Athens metropolitan area comprises of 57 municipalities (including the municipalities of Athens and Piraeus, the port of Athens) and has a population of approximately 3 million people.

Sample selection

For the initial sample selection a private company, specializing on opinion polls was hired. They visited 2,000 randomly selected homes within the metropolitan area of Athens and collected from each subject a two page questionnaire regarding their age, sex, occupational, socioeconomic and educational status, and whether they were smokers or not. Out of these 2,000 individuals a little more than half were smokers. All questionnaires were completed and handed over to our lab during May and June of 1996.

For the *Exposure sample* subjects with the exception of five persons, only non smokers were used. Individuals were selected randomly out of the large sample and contacted by phone to be asked to participate in the study. Those that did not agree to participate were replaced by the individual corresponding to the next random number. Overall, out of every ten to fifteen individuals called one would agree to participate.

For the *Diary sample* subjects again with the exception of five persons, only non smokers were used. Initially subjects were contacted by mailings. Every few months 200 letters containing a questionnaire, a TMAD, instructions on how to complete them and a letter from our lab explaining the purpose of the study were mailed out. The response rate was in the order of seven individuals per mail out, so by the time all non smokers had been contacted there were still approximately 15 subjects missing. For the remaining people the same approach with the exposure subjects was followed: the individuals were contacted by phone and if they agreed an *EXPOLIS* researcher would visit their home.

Exposure subjects

The field work with Exposure subjects was completed in two phases. Phase one started on January 17, 1997 and lasted till July 4, 1997. Phase two started on September 9, 1997 and ended on March 17, 1998. Twenty three subjects were measured during phase one and 27 in phase two. With the exception of the two summer months, when a large number of citizens move away from the city and thus traffic conditions are not representative of the rest of the year, the field work covered all seasons.

Out of the 50 subjects a work MEM was placed at the workplaces of twenty nine. Out of the remaining 21, eleven were working at home, five were not working in a specific location, four were students and one could not get permission from his employer.

The PEM pump worked without any problems with all of the subjects. The only problem encountered was when a subject forgot to change from day to night filter. A few days later the subject was given the PEM case again together with a new copy of the TMAD and the Short-Term Recall questionnaire and the measurement was repeated.

The CO monitors failed with four subjects, in all cases due to download problems after the completion of the sampling. Again measurements were repeated, with the subjects carrying only the monitor along with a new copy of the TMAD and the Short-Term Recall questionnaire.

Finally, the MEM pumps failed with six subjects. In all cases it was either the home indoor or home outdoor pump. The reasons were that either the maximum load on the filters was exceeded or the battery run out of power. In all cases both the home indoor and the home outdoor pumps were placed again at the subjects home a few days later.

4.2. Basel

Population sampling

We could obtain the primary random sample from the civil register of the city of Basel without problems because of the scientific aims of the *EXPOLIS* study. As one third of the population of Basel are of Non-Swiss nationality, partly not speaking German, we expected that a part of our target population would not be able to participate in *EXPOLIS* due to language problems. However, as it was not possible to predict a person's mother tongue from the Nationality, no specific Nationality was excluded a priori. In the random sample of 3'000 subjects, 66 nationalities were represented, with Italians (7.4%), Turkish (4.3%), Spanish (3.5%), Germans (2.9%) and Ex-Yugoslavians (2.8%) being the largest groups besides Swiss nationals (68.6%). As expected, the response rate (valid short screening questionnaire) of Swiss Nationals (57.4%) was clearly higher than of Non-Swiss Nationals(29.3%). Whether this lead to a selection bias towards more or less exposed participants will be further investigated, using traffic volume at home (cars/h, lorries/h) as exposure proxy, which is available for 2582 subjects (86%) of the random sample.

Field Work Experiences - Feasibility of Personal and Microenvironmental Measurements

Pre-Pilot study in Summer 1996

In Basel, a Pre-Pilot study was conducted in July and August 1996 with four participants. Feedback of participants showed clearly, that the MEM-equipment was still too noisy for running at workplaces and insides at home. We evaluated several insulation materials and found a noise-absorbing, self-adhesive foam rubber, 13mm thick, which reduced the noise level of the operating MEM-equipment significantly when the inner side of the MEM box, containing the PQ100 pump, was fully covered with it.

The expected PM2.5 concentrations imply a weighing precision of 1 µg. Despite the use of a Micro-balance, first weighing tests with Blank filters showed random variations of 10 µg, consistent with experiences in Helsinki. Using Polonium 210 for deionising the teflon filters as proposed by *EXPOLIS* Helsinki, was not feasible in Basel. However, with the Haug Multistat Ionizer (Multistat Haug AG), an equivalent alternative was found to solve the problem of electrostatic charges on teflon filters.

Because deionising had not yet been implemented and the bubble metres for flow measurements before and after MEM and PEM sampling had not yet been available, PM2.5 concentrations were not calculated for the pre-pilot.

Pilot study in Winter 1996

Five subjects were enrolled for the pilot study in December 1997. The major problem, showing in this stage, was the leaking PEM filter holder. After implementing the instructions of Helsinki (sealing the 37mm filter holders with scotch tape), the PEM measurements were reliable and the main study could be started in February 1997.

Main study

The fact that all 50 subjects who were enrolled for the main study in Basel successfully completed the study protocol, shows, that the personal and microenvironmental measurements as carried out in the *EXPOLIS* study are feasible, though very time consuming for the research teams. Only 3 subjects forgot to change the PEM filters at the right moment, so that PM2.5 „private“ and PM2.5 „workday“ levels cannot be distinguished. Several participants stated, that they would rather not continue longer than the 48-hour measurement period, because the PEM case was too heavy and interfering with their daily activities such as shopping, going to the cinema or outdoor sports (e.g. jogging). Thus, for obtaining good compliance also for repeated personal measurements on the same subject, the personal measurement equipment should be designed rather lighter than the 5.2 kg PEM-case used in *EXPOLIS*.

VOC analysis with an alternative method

(*Charcoal instead of Tenax*)

Financing of VOC analysis in *EXPOLIS* Basel could only be obtained for the collaboration with a local partner. No Swiss laboratory was ready to perform VOC analysis with the required *EXPOLIS* technology (Tenax Tubes). However, Carbotech AG (Basel) agreed to adapt their technology (Charcoal tubes) to the PEM and MEM equipment used in *EXPOLIS*. The comparability of the two methods was assessed in 2 Inter-calibration tests organised by JRC-EI and seven 24-hour MEM parallel measurements in Helsinki. The major drawback of using the charcoal tubes in *EXPOLIS* Basel was that ten of the *EXPOLIS* target compounds could only be determined qualitatively and for five compounds the method was not suitable. 16 compounds could be quantified.

Overall, the experience made clearly showed, that within a scientific multicentre project, use of one common method and laboratory for all centres would be the preferred way to go. Intermethod comparison, a prerequisite for sound scientific data interpretation, is unpredictably costly and time consuming and the risk of getting uncomparable data is high. It is to emphasize that this is not a statement in favour or against any of the methods or laboratories but rather to the general approach of selecting different laboratories. The collaboration with the local CARBOTECH laboratory was excellent and driven by achieving the highest quality standard possible.

Research collaboration in Basel

Throughout the *EXPOLIS* project, we had close collaboration including regular meetings with the BRISKA project team (PI: Charlotte Braun-Fahrländer). The Basel Air Pollution Risk Study is funded by the interdisciplinary MGU Program of the University of Basel. It measured a wide range of particulate matters and other pollutants (such as PAH, VOC, NO2, EC / OC) at six fixed sites of diverse locations in Basel with repeated continuous measurement periods of 13 days each. The project will allow to assess spatial distributions of ambient pollutants to apply in risk assessment. The two projects will strongly profit from each other as they where conducted simultaneously. BRISKA enriches the analytic options of *EXPOLIS* Basel data with regard to offering a broader range of pollutants measured at fixed sites. Furthermore, it is trough this collaboration that the second *EXPOLIS* project, the Elemental Analysis Study (EAS-*EXPOLIS*) will be able to analyse the chemical elements on the collected PM2.5 filters. BRISKA and EAS-*EXPOLIS* will strongly improve the source apportionment of particulate pollution in Basel. The Swiss part of EAS-*EXPOLIS* is funded by the Swiss National Science Foundation in will last to 2001.

4.3. GRENOBLE

In Grenoble, the field work was first divided into 2 phases, a « summer » phase (1996) and a « winter » phase (1997), to assess the effect of seasons on exposure levels. There is an additional ongoing phase this summer 1998. Within a research program on air pollution and health, a study was carried out in parallel to *EXPOLIS*. It aimed at assessing the short term effect of air pollution exposure on a panel of asthmatics and controls. As a result, the recruitment in Grenoble differed from that of other *EXPOLIS* centres. Volunteers were recruited through the Grenoble Hospital Pneumology service (Prof. C. Brambilla) and the local press (« Dauphiné Libéré »). This process was successful and most of the « exposure volunteers » were recruited, based on the *EXPOLIS* criteria, within 2 weeks after the papers were published in the press. However, due to lack of funding, it was not possible to extend the study to a large sample of « diary only volunteers ».

Because of the specific interest of local sources of funding (French car industry: Union Routière de France), it was decided to design the study in order to be able to compare exposure associated with traffic emissions and total daily exposure. The exposure assessment study was slightly modified accordingly. Instead of one single monitor per PEM-case, 2 were used. They allowed direct personal PM 2.5 measurements both for total exposure (48 h. cumulated) and for indoor exposure. Then, outdoor personal exposure could be obtained by computing the difference between the 2 measures. To do so, each volunteer was asked to put on hold one of the pump each time he(she) spent more than 5 minutes outdoors, including while commuting (car, bus, tram...). Hence, this first pump was only working while indoor, collecting « indoor » particles. The second pump ran continuously during 48 h. and the filter collected particles while indoor and outdoor.

The aim of this procedure was to assess the part of the personal exposure to fine particles attributable to urban traffic. No micro-environmental measurements were carried out because of funding limitations. However, a local collaboration (Prof. Masclet, ESIGEC, Université de Savoie) allowed us to perform preliminary elemental chemical analysis on some of the Grenoble personal filters in order to characterize which pollutant(s) adsorbed on fine particles best indicated motor vehicles as the source of urban air particulates. Total carbon has been measured by aethaelometry. The 16 main PAHs according to the EPA list for carcinogens were measured by HPLC. In a further additional study, heavy metals might be quantified for a few filters using the PIXE technique (responsible : H. Guegan, CENBG, Bordeaux) in the framework of a French Ministry of Environment funding : PREDIT-PRIMEQUAL.

Moreover, in addition to the *EXPOLIS* questionnaires, each volunteer filled in a map with all his(her) commuting along the 2 days of personal measurements (road, time, distance, length). Using the traffic intensity data retrieved from the transport authorities on each of the streets that were used, an index of exposure to traffic was computed. Its rationale was to weight the time spent on a given road by the traffic intensity at the time the volunteer was on that road, summed up over all the daily transfers. Using these data, this index will be included as a variable in the model that will try to evaluate the part of urban traffic exhausts in PM 2.5 personal exposure. In addition to exposure while commuting, traffic emissions contribute to personal exposure at the place of residence, of work and of other daily activities. As a first estimate, the influence of traffic emissions on these indoor environments will be assessed through the distance between these locations and the nearest heavy traffic road. For this purpose, a Geographic Information System will be used.

Another important focus of the Grenoble study is the assessment of the validity of fixed ambient air monitors as a measure of population exposure to air pollutants. Results from the Grenoble Air Quality Monitoring Network (ASCOPARG, M.B. Personnaz) were collected in parallel to the *EXPOLIS* personal measurements. Like in most European towns, only Black Smoke and PM 10 data are measured by fixed monitors. PM 10 are measured using a Tapered Element Oscillating Micro-balance (TEOM) on 2 urban sites : one is a proximity site (situated on a high traffic level street, with a high NO/NO2 ratio) ; the other one is a background site (very low traffic, low NO/NO2 ratio). In order to convert PM 10 into PM 2.5 values, a GRIMM (G1105) was used to describe the granulometric profiles of particles on both sites. Measurements were made during the summer and winter periods. Associations between PM 2.5

personal exposure and PM 10 ambient air values during the same days were assessed in multiple linear models where important covariates were accounted for (ETS, residence floor...). This study aims at weighing the values provided by, respectively, the background monitor and the proximity monitor, in order to estimate the *EXPOLIS* personal exposure.

Dealing with the PM 2.5 mass determination, the usage of a deioniser was not part of the Standard Operating Procedures (SOPs) during summer 96. As a result, all the weight measurements performed during this first phase were invalid. To face this problem, the PM 2.5 masses were evaluated using the reflectometry technique (optical determination, based on the OECD Black Smoke references). In winter 97, both deionised weighting and reflectometry data were available. A third phase (June-July 1998) was organized in order to obtain some summer PM 2.5 deionised weight measurements, thus allowing comparison of the data between winter and summer. During this additional phase (concerning 20 volunteers), CO is also measured (following the *EXPOLIS* SOP) using Langan monitors, along with some aldehydes badges (GMD 570) that are tested on 10 volunteers. This badges will be analyzed by the GRECA (Groupe de Recherche en Environnement et en Chimie Appliquée),V. Jacob, Grenoble) using the HPLC technique. In parallel, each volunteer fills in the *EXPOLIS* questionnaires. VOCs and CO were not measured in the other phases, also for financial constraints.

This project would never have been implemented in Grenoble without the technical and financial support of 2 institutions: APPA (Agence pour la Prevention de la Pollution Atmosphérique ; responsible in Grenoble J.Déchenaux) and ADEME (Agence de l'Environnement et de la Maitrise de l'Energie, responsible for the project : F.Dor). Many other parties also brought their support: the French Ministry of Environment, the Grenoble Communauté de communes and the Rhône-Alpes region. Help from the Isère département authorities is also acknowledged. It should be emphasized that this project did have and still has some impact among the general population in France. Local newspapers and TV channels, but also National media are interested in this study which began just after the publication of the French Air Quality Act by the Ministry of Environment.

4.4. HELSINKI

The *EXPOLIS* study started in Helsinki in March 1996 when the field workers were hired to the project. The Helsinki group was responsible for the coordination of the project starting from equipment delivery and training programme development. In the beginning of the project five PhD. students were hired for this work. The first job was to deliver and test all the equipment needed to develop feasible monitoring packages for personal and microenvironmental measurements for $PM_{2.5}$, VOCs, CO and NO_2. It proved to be a very time consuming to get all the monitors and procedures to function properly. After three months of delivering and testing the monitors the first pilot subjects were measured in June 1996.

Pilot phase

From June to August 1996 13 subjects were measured in Helsinki to practice procedures and to test monitors and work procedures in the field. In the following a list of some typical researchers' experiences from the field work is presented:

- problems how to explain the subjects that I can not take her to my car after the set up visit to her home before going to set up work monitor (subjects should not change their commuting type during measurement period)
- interview (questionnaire) couldn't be finished, because the subject was not at home, when visiting him after the sampling period. (his son let us in)
- a simple minded person, but there were no problems in handling PEM-filters

- study is very strenuous for the subject. 48-hours is the maximum time for loading of the subject
- the early Monday morning visit to subject is a problem (people are too busy)
- car parking is an issue. Where can you leave your car in downtown when visiting subjects
- working days for researchers during the field work can be very long (from early mornings to late evenings)
- subjects are easy to contact
- mostly they are very co-operative
- need to check carefully the filled in forms after the measurement- often there are little things missing
- some subjects said that two days is the maximum time to support the measurements because of the noise, the trouble in keeping track of comings and goings etc.
- some subjects found that the PEM case restricted their life. As a result the case was either not carried along all the time or the subject stayed home more than usually.
- rucksack was needed for PEM when the subject needs to have his/her hands free when shopping or carrying bags
- one subject did not take the PEM-case to the bedroom
- one subject did not open the windows as usually
- PEM-case was too heavy (instead of walking used a bus)
- the case was too noisy
- one subject did not want that somebody could notice the PEM case
- there was a mistake in changing filters; filters 3 and 4 where used conversely
- VOC-calibration - 1 mL/min - could be painfully slow
- MEM box is heavy to carry i.e. to the fourth floor (without a lift), Isn't there any better procedure for that?
- problems with programming PQ100 pump (Indoor MEM didn't run the 2nd night)
- about one year old child was interested in home MEM
- subject's cat chewed the hose of the VOC-tube
- filter weighing is problematic
- MEM duplicate filters were mostly ok, but in 10-30 % of duplicate measurements there was notable discrepancy. This must be solved
- PEM duplicate filters did not work at all (due to weighing problems)
- some questions are not 'waterproof' like the amount of disturbance, distance of house from road
- the database is very helpful in printing out different types of results
- data entry tools must be developed; the plain tables/queries are not very easy to use
- data coding must be fixed and trained to every user. This is most important with class variables, where the increment/decrement of one class might totally change the meaning of the value

The most serious problem according to pilot phase findings was weighing procedure. Variation in the weighing results was too high. The reason for that was static charge in the filters. This problem could be avoided by deionising the filters with a PO-210 alpha emitter. After starting to apply the deioniser, weighing repeatability was very good. After solving the weighing problems there were still problems with PEM duplicates. After careful testing we found a leakage from most of the PEM filter holders. It was caused by incomplete tightening of the filter holder parts after weighing. This problem was solved by tightening the parts carefully and taping the sealing between filter holder parts.

After the pilot all procedures were finalized and after that the quality assurance tests were repeated. The results showed good repeatability for the measurements and thus actual exposure measurements could be started.

Field phase

In the field work there were 5-7 researchers for about one year in Helsinki. They could measure 201 subjects and collect questionnaire and time activity diary data from 234 additional subjects in that time. Thus field work is time and researcher consuming in this kind of project, where personal and microenvironmental measurements are carried out.

In Helsinki study subjects were co-operative and motivated. We did not loose any monitors or researchers during the field work, which we were afraid of beforehand.

Overall summary of the experiences

During this study we have met a lot of study subjects, which has given us new experiences and maybe taught us new social skills. We have understood the importance of quality assurance work, we have met colleagues in other *EXPOLIS* centres and had very good co-operation with them and at last but not least we had several Workshops around Europe where we had nice time together with our *EXPOLIS* society. In those Workshops we were exposed, besides hard work, to dancing, skiing, nice dinners, and everything else, which leads, according to our hypothesis, to harmless medical and mental end points. In EXPOLIS our duty has been to build up a house with Czech, Dutch, Finnish, French, Greek, Italian, and Swiss bricks. Now this house has got its shape, but the size of the house in not yet known. The house size depends on, besides our work, the visitors who might have new bricks and new tools. We are looking forward to visitors.

4.5. Experiences in Milan

In a previous study made from July 1995 to July 1996, the Milan group evaluated the daily personal exposure to PM10, NO2 , CO, Benzene, Toluene and TVOCs of a hundred office workers living in the metropolitan area of Milan (Carrer, 1997); the relative contribution of office, home and transport means to the daily exposure was also assessed. The assessment of the daily personal exposure to the pollutants was performed with sampling sessions at the office during working hours, inside the home during the participant's permanence, and during commuting. Information about the microenvironments and the activities of the subjects was collected through a home-office-transfer microenvironment questionnaire and a time-activity diary. This study has been an useful experience for the setting up of the daily personal assessment and microenvironment questionnaire and time-activity diary in the *EXPOLIS* project.

For the *EXPOLIS* study in **Milan**, the *Diary sample* was based on a random draw from the city inhabitants; the *Exposure sample* was selected from office workers of public and private buildings located in Milan. The *Diary sample* (250 subjects) was formed stepwise by a random draw of 3009 adults (25 - 55 years of age) obtained from the Municipal Civil Register of Milan. A short questionnaire was mailed to this base population sample; the final *Diary sample* was drawn randomly from the base sample after having excluded the unwilling or unqualified individuals. Each selected subject was personally contacted to fill in the questionnaire and the time-activity diary. All the subjects were contacted by the same researcher, about 20 subjects per month from June '97 to May '98. The data were recorded in the *EXPOLIS* database.

In Milan, since over 75% of the working population operates in offices or similar microenvironments, it was decided to evaluate the exposure for only this category of workers. The Exposure sample (50 subjects) was selected from office workers of public and private buildings located in Milan; these

buildings had been previously evaluated by our Institute in former studies and can be considered representative of the different building typologies.

The measurements were performed from March '97 to January ’98 (about 6 subjects per month).

The EXPOLIS SOPs (Standardised Operations procedures) were followed even if some little changes have been applied.

In measuring the CO exposure, the monitors used were already present before starting the activities; since they did not have appropriate technical characteristics to measure the external temperature this was omitted. Temperatures were although measured with the internal sensor (channel 3) and recorded in all subjects. The download software used was the original Eibearm software supplied by the manufacturer, Langan, and three files were created for each subject. At the end of field activities these three files were combined in one for each subject (channel 1, 2, 3).

The VOC measurements have been performed following the SOPs and the samples were analysed in the VTT laboratory. The shipment procedures were organised to optimise the workload of the laboratory according to the laboratory staff. The calculating activities were performed in a separate session in accordance with the SOPs by a member of the Italian team after receiving all chromatograms and relative reports from lab. Blanks and duplicates were collected as the number previously determined.

The PM measurements were performed following the SOPs, but the deioniser was not used in any case for Department security rules (use of Polonium and/or high voltage instruments is forbidden). In each case 4-6 sequential weighting operations were performed until the weight was considered stable before and after the sampling activities.

All the PEM and MEM sheets were properly filled and all the microclimatic parameters were recorded, also in the weighing room. The MEM programming with “fake time” presented some troubles especially in the first phase of measurement.

4.6. Experience and comments from Prague

A specific problem in the Prague study was the selection of persons who would agree to participate in the study. Inhabitants of the studied areas were informed about the study through daily as well as regional journals. Nevertheless, the interest among inhabitants to participate in the study was very small and the number of returned introductory questionnaires was very low. It also happened that some subjects, who originally agreed to participate in the study, refused to collaborate when they were really contacted. Moreover, in the course of the study we had also problems to establish a contact with some of the selected subjects; there was a long time lag between the registration of the selected subjects and the realisation of our measurements, and in the meanwhile some subjects either moved or changed their employment. In the case they gave only one contact address (home or employment address) which was changed, they had to be excluded from the selected group.

Many more individuals were willing to participate in the measurements than in completing the questionnaires, although the participation in the measurements brought them certain discomfort. The probable reason was that they were curious about the values of pollutants in their environment. Judging from the contacts with the registered subjects they were mostly individuals with a higher than average degree of education and socioeconomic level. It also explains their interests about the environment and life-style, which is confirmed e.g. by the low number of smokers in this group.

On the other hand, we can say that all subjects who participated in the study were very co-operative and had a great interest in the results of the measurements and their evaluation related to the possible effects on their health. The employers had also similar interest in the results concerning the measurements of the working environments.

Comments of the subjects carrying the PEMs: the stories about the behaviour of other people was very interesting, sometimes amusing. For example, one PEM invited a great attention in the Metro, in a theatre or restaurants, the PEM filled some people with suspicion that it is a dangerous thing. The PEMs were carried mostly in rucksacks or handbags. Due to its weight the PEM was a burdensome load especially for women, who had to go shopping after their work. Similarly, the PEM was uncomfortable for women in their business rounds. That is why they preferred to carry the PEM on their back, but it was not realisable in some situations (business meetings, theatre, etc.). For men the PEMs did not represent such a great load, they had better good physique and drove more often by car.

Filters exchange did not make serious problems, nevertheless, some mistakes occurred; only one person was not able to exchange the filter although he had detailed instructions and explanation.

Due to the higher noise level of MEMs they were preferably located in living rooms, where their operation interfered less than in bedrooms.

In the course of study (middle of the period studied) we had some difficulties with the program, especially with automatic switching off the MEM pumps. The wear of some components of the MEM was the reason. Similarly, some technical defects were found in the CO monitors and therefore some measured data from some subjects were lost. It was found that it is not good to manipulate excessively with Langan in the laboratory after preparation of PEM to measurements - it can damage contacts. To reduce the problems with the CO monitoring (drop-out) we checked Langan also immediately before departure to the examined person.

We can conclude, that our participation and co-operation in this study was very interesting for us bringing experience not only in contacts with the subjects but also in co-operation with our foreign partners and colleagues. I think, there are no comparable data in our Republic obtained in similar way and the conclusions of this study will represent a great contribution to environmental studies on national level and also for international comparison.

5. RESULTS

The following presentation of the results are based on cleaned and final data, but present only some preliminary overviews and analyses of the very large database that has been collected in the EXPOLIS study. The results are organised according to the data collection tools and mostly presented in a collection of data tables in Annex II. A great deal of these tables present questionnaire results from the 6 studied cities, which has been collected as background data for future exposure modelling and simulation needs, but which may also be of interest to researchers concerned with time activity patterns, household and workplace characteristics, equipment and activities across European cities. These preliminary results are subject to changes due to critical evaluations within and outside of the EXPOLIS teams. Further analyses will be presented in scientific conferences and submitted for publication in peer reviewed scientific journals. **Chapter 6.** Discussion will discuss the data in the context of the study goals.

5.1. Short Screening Questionnaires

Between and within centres comparisons of the Base population samples v.s. the Diary samples v.s. the Exposure samples

Annex II: Table 1. Presents the overall numbers of approached and completed subjects of the different samples. In Helsinki the questionnaires were first mailed and then remailed to those, who did not reply to the first mail. Finally telephone interview was applied to those, who did not respond to two mailings. This three step procedure, plus the fact that Finnish citizens are known to respond exceptionally positively to population surveys of clear public interest, explain the high overall response rate (75%) in Finland. In other centres door to door interviews and mailed questionnaires were both applied with variable, 5% (Prague) ... 49% (Basel), response. In Grenoble, no separate Base sample was formed. The response rates of the Diary and Exposure samples were much higher, generally 100 %, because the individuals were approached personally, and those who would not comply - by accepting the equipment to their homes, workplaces and personal monitoring (Exposure sample), or by arriving to the instructing session or accepting a field staff member's visit for instructions - were not included in the sample.

Annex II: Tables 2. /A...P present the results of the short two page questionnaires from each EXPOLIS centre. As these questionnaires were applied at a very early stage of EXPOLIS cooperation and teams training, regrettably the questionnaires were not completely identical in each centre. Basel, Grenoble, Milan and Prague share a similar questionnaire, Helsinki and Athens - starting the work before the other centres - used somewhat different short questionnaires. Direct comparisons between the cities must therefore be limited to the questions that were the same in the centres to be compared, or to the questions where the answers are otherwise known (e.g. no metro in Grenoble, no single glazed windows in Helsinki).

Because the subjects to be approached in each centre for the *Diary* and *Exposure samples* were randomly drawn from the individuals that had completed the short questionnaires for the *Base sample*, the three samples in each centre can be directly compared to each other to evaluate what if any selection biases were caused by the rather involving requirements for the *Diary sample* subjects (long questionnaire, 48 h recall questionnaire, and 48 h Time-Microenvironment-Activity-Diary) and the quite invasive procedures and requirements of the *Exposure sample* subjects (personal and microenvironmental monitors, filter changes etc.). This direct comparison, however, cannot be made

for the smoking related questions, where the proportion of smokers in the *Exposure sample* was limited to 25% or less in order to avoid the results to be overwhelmed by the effects of smoking, and consequently to contain little information of any other sources of air pollution exposure. The *Base sample* should be representative of the proportion of smokers in each city.

The (other than smoking) significant (p # 0.05) differences between the *Exposure sample* and the *Base sample* were found for occurrence of asthma, time spent outdoors and student status in Basle, for workplace, use of train or metro and occupational class in Helsinki, for number of adults in household, workplace (due to specific study design), use of bus or tram and occurrence of wheeze (asthma symptom) in Milan. In other centres the *Base samples* were either too small (Grenoble) or too unrepresentative (Prague) for conclusions, or no significant differences were found (Athens).

Annex II: Table 3. presents some comparisons between the centres. The *Base sample* individuals appear to be rather evenly distributed between the three age groups, 25-35, 35-45 and 45-55 years, with Helsinki having the oldest sample, and Prague the youngest. Both genders are well represented in each centre. The proportion of smokers is highest in Athens, 52%, followed by Milan, 36%, Basle, 34% and Helsinki, 28%. In Grenoble, only non smokers participated and in Prague the response rate is too low for comparison. The proportion of single adult households was highest in Basle and Helsinki, lowest in Milan and Athens. More than 50% of the responding urban households in each centre had no children at home, in Grenoble, Basle and Milan this portion was over 65%. For commuting to work in winter, over 40% used their own cars in Grenoble, Helsinki and Milan, only 20 % in Basle and 12% in Prague. Athens data are missing.

5.2. Core Questionnaires

Core questionnaires were applied to all subjects in the *Diary* and *Exposure samples.* The total numbers of individuals, see Annex II: Table 4., in the *Diary* and *Exposure samples* were relatively large in Helsinki (both over 200), Basel and Milan (large *Diary samples*), and smaller in Athens, Grenoble and Prague (small *Diary and Exposure samples*).

Home Environment; Annex II: Tables 5. /A...P

These Annex II: Tables describe the environments of the homes of the subjects in the *Diary* and *Exposure samples*.

Comparing the Centres:

The proportion of downtown residents in the *Diary* and *Exposure samples* was extremely high (80%) in Prague, 34-40% in Milan, Grenoble, and Basel and less than 20% in Helsinki and Athens. Most (from 58% in Helsinki to 98% in Milan) live in multistory apartment buildings. The proportion of subjects living in buildings built before 1970 was high, 86%, in Basel and Prague, low, 26 %, in Athens and, 37%, in Helsinki, with Milan, 72%, and Grenoble, 51%, falling between the extremes.

Men vs. Women:

The only statistically significant difference between the men and women was the reported (subjective) traffic density on a nearby street in Grenoble, Milan and Basel. In Athens and Helsinki the men and women reported quite similar traffic densities.

Diary and Exposure Samples:

Significant differences were observed in Athens for *built year* (more *Exposure* than *Diary sample* subjects lived in quite new dwellings), in Helsinki for *traffic on nearby street* (more heavy and less light traffic reported by the *Exposure sample*), in Milan and Prague for *home location*.

Home Description; Annex II: Tables 6. /A...P

These tables present the statistics of the home building and furnishing materials, basic equipment and the presence and quantity of smoking at home.

Comparing the Centres:

Among the potentially problematic home materials, wall to wall carpets in some rooms are common in Prague (72%), Basel (61%) and Grenoble (51%) but infrequent (<20%) elsewhere. Chipboard as wall material is present in 23% of the homes in Helsinki, 18% in Milan and 14 % in Basel, but very little elsewhere. In the climate of Helsinki double glazed windows are the minimum (triple is the standard in post 60's buildings), but they are quite common all over Europe - from 33 % in Athens to 72 % in Prague.

Central or/and gas heating systems dominate in Athens, Basel, Grenoble, Milan and Prague. District heating heats 75% of the homes in Helsinki and 33% in Basel. Electric heating is common in Grenoble and Prague, 21%, Helsinki, 15%, and Athens, 13%. Fireplaces can be found from 21% of the homes in Athens, 12% in Helsinki and less than 8% elsewhere.

Air conditioners are quite common in homes of Athens, 46%, common also in Milan, 11%, but very infrequent elsewhere. Air humidifiers are common in Basel, 20%, and Milan, 11%, but quite infrequent elsewhere. Air cleaners or ionisers can be found in 11% of the homes in Helsinki, 8% in Prague, but only 0-4% elsewhere.

Gas cooking dominates in Milan, 98%, Prague, 62%, and Grenoble, 58%, is also common in Basel, 37%, but much less used in Athens, 10%, and Helsinki, 6%. Cooking with a solid fuel fired stove is a rarity (#1%) in each of the *EXPOLIS* cities. In Prague 64% of the kitchens are not equipped with kitchen fans (filter and recirculation) or extractors, in Basel 35%, in Grenoble 18%, in Athens 15%, but in Milan and Helsinki over 90% of kitchens have recirculating fans or mechanical extractors.

The prevalences of smoking - except in the *Diary* and *Exposure samples* - should not be studied from these tables because the numbers of smokers are limited by the sampling procedure.

Men vs. Women:

The statistically significant differences between the home characteristics of men and women in the *EXPOLIS* samples are few. In Basel more men than women live in homes with plaster board walls, In Grenoble more women than men have gas stoves, in Milan women have more soft furnishings, less kitchens with fans, men have more wooden panels. No significant differences between the home characteristics of men and women in the *Diary* and *Exposure samples* were detected in Athens, Helsinki or Prague.

Diary and Exposure Samples:

The statistically significant differences between the home characteristics of the *Diary* and *Exposure samples* are also few. In Basel more subjects in the *Exposure sample* had chipboard walls, and while four subjects in the *Exposure sample* were heating their homes with coal fire, there was only one in the *Diary sample*. In Helsinki more subjects had central heating systems in the *Exposure sample*, humidifiers or electrical air cleaners in the *Diary sample*. In Milan curtains were more common among the *Diary sample*, plasterboard walls and wallpapers among the *Exposure sample*. Again no statistically significant differences were observed in the Athens, Helsinki or Prague samples.

Workplace Environment; Annex II: Tables 7. /A...P

These tables describe the environments of the workplaces of the subjects in the *Exposure* and *Diary samples*.

Comparing the Centres:

The location of the workplaces of the *EXPOLIS Exposure* and *Diary sample* subjects in the downtown area is most pronounced in Milan, 65%, followed by Prague 58%, Helsinki 53%, Grenoble , 52%, Basel 50% and Athens with only 32%. Office buildings are the most common work environments, from 75% in Prague to 38% in Basel. Industrial buildings follow in order from 20% in Basel to only 3% in Prague. The youngest workplace buildings are found in Athens, only 29% built before 1970, 25% after 1980, and Helsinki, 40% before 1970, 34% after 1980, while the oldest workplace buildings were in Milan, with 59% (Basel, 57%, Prague 55 %) built before 1970. In Grenoble 62% and Athens 54% of the subjects reported (subjective perception) heavy traffic on a nearby street, while in Milan only 41% and Basel 44% reported heavy traffic. Helsinki and Prague fell between the two groups.

Men vs. Women:

The statistically significant differences between the workplace environment of men and women in the *EXPOLIS* samples are few. In Basel, Helsinki and Milan significantly more men work in the industrial area (and buildings) and women in the downtown area (and office buildings). Also in Prague and Athens more women than men work in the downtown area and office buildings, but the differences are usually insignificant due to small sample sizes.

Diary and Exposure Samples:

Only in Milan can statistically significant differences be found between the workplace environments of the *Exposure* and *Diary samples*. The exposure sample is much more concentrated to the downtown area, and working in pre 1970 built office buildings. This is due to the specific sampling design in Milan.

Workplace Description; Annex II: Tables 8. /A...P

These tables present the statistics of the workplace building and furnishing materials, basic equipment and the presence and quantity of smoking.

Comparing the Centres:

Among the potentially problematic materials, wall to wall carpets at the workplace are the most common in Prague (70%) and Basel (57%), common also in Athens (29%), Grenoble (25%) and Milan (19%), but quite rare (6%) in Helsinki. Chipboard as wall material is present in rather similar concentrations all over Europe, ranging from 23% in Athens to 13% in Prague. In the climate of Helsinki double glazed windows are the minimum (triple is the standard in post 60's buildings), but they are common all over Europe - from 18 % in Athens to 72 % in Prague.

Central or/and gas heating systems dominate in Athens, Basel, Grenoble, Milan and Prague. District heating heats 88% of the workplaces in Helsinki and 34% in Basel. Electric heating is common in Athens, 29%, Grenoble, 22%, and Prague, 16% and less frequent elsewhere. Fireplaces can only be found in #1% of the workplaces in any of the *EXPOLIS* cities.

Air conditioners are quite common in the workplaces of Milan, 64%, Athens, 62%, Grenoble and Helsinki, 45%, and rather common also in Basel, 24%, and Prague 19%.

The prevalences of smoking - except in the *Diary* and *Exposure samples* - should not be studied from these tables because the numbers of smokers are limited by the sampling procedure.

Men vs. Women:

The statistically significant differences between the workplace characteristics of men and women in the *EXPOLIS* samples are the following: In Athens more women have curtains at their workplaces, and more men work in district heated buildings. In Basel more men work in buildings with double glazed windows. In Grenoble more women work in buildings with district heating, soft furnishings and wallpapers, and more men in buildings with electric heating, double glazed windows. In Helsinki more women work in buildings with curtains and chipboard structures, men have more often humidifiers. In

Milan women work more often in buildings with wall to wall carpets, soft furnishings, and wooden floors, while men have more often air conditioning, humidification and electrical air cleaning. No significant differences between the workplace characteristics of men and women in the *Diary* and *Exposure samples* were detected in Prague.

Diary and Exposure Samples:

The statistically significant differences between the workplace characteristics of the *Diary* and *Exposure samples* are few. In Basel more subjects in the *Exposure sample* had air humidifiers in their workplace. In Helsinki three *Exposure sample* subjects had fireplaces at their workplaces vs. none in the *Diary sample*. In Milan, curtains, wooden floors and plasterboard wall were more common in the *Exposure sample*. In Prague, plasterboard walls were much more common in the workplaces of the *Diary sample*.

5.3. The Short-Term Recall Questionnaire

This questionnaire was used to collect retrospective data about potentially exposure affecting events or activities that had occurred during the 48 hour period when the exposure and microenvironmental samples were collected and the Time-Microenvironment-Activity-Diary was filled. While the core questionnaire asked about the presence of certain equipment, the recall questionnaire asked about their use or operation during this time.

Frequencies of Equipment Use and Activities at Home and Workplace;Annex II: Tables 9./A...P

In these tables the question answered is how many of the *Exposure* and *Diary sample* subjects had engaged in the activities of concern, during the 48 h follow up period, irrespective of frequency or duration.

Comparing the Centres:

At home the gas stove was used the most in Milan, by 87% and in Prague, by 65% of the subjects. In other centres the gas stove use percentages reflected the availability of gas stoves and were 40% or less. Kitchen fan was used by 53% of the subjects in Athens, about 40 % in Basel, Helsinki and Milan, but only 17% in Prague and 6% in Grenoble.

Other gas appliances were either nonexistent or were used very little. Vacuum cleaners were used during the 48 hours rather evenly across Europe, most in Milan, 61%, least in Grenoble, 27%, with other centres between 36 and 45%.

The use off supplementary heating equipment was quite infrequent, except for the use of gas heaters in Prague by 30%, Grenoble, 19%, and in Milan, 5% of the subjects.

Windows were opened for ventilation by 86% (Helsinki) to 100% (Grenoble) of the subjects. Because the sampling was distributed around the year, the use of air conditioners was much less frequent ranging from 15% in Athens to 0% in Grenoble. In spite of the fact that relatively many households had air humidifiers and electric air cleaners, they were used extremely little, in only 0...3 % of the households. Wood heated sauna was used in 4% of the households in Helsinki (most saunas are electric), and one (!) household in Basel.

In the workplace the use of any form of supplementary heaters was limited to a few % in any centre. In the workplace windows were opened less (from 43% in Helsinki to 76% in Prague) and air conditioning used more (from 11% in Prague to 38% in Grenoble) than in the homes, obviously due to more busy (and polluted) location in the city, less individual ventilation controls and access to the window. The use of humidifiers and air cleaners was limited to less than 10% in each sample.

Photocopiers and laser printers are among the most common work equipment today, and they were used by, or operating in the same room of 21% (Prague) to 100% (Grenoble) of the subjects of this study.

The most common exposure related activities were using deodorants and perfumes (from 92% in Milan to 71% in Athens - only 40% in Grenoble, where the asthma status of the subjects affected their behaviour), using cleaning chemicals (from 63% in Athens to 35% in Basel - 16% in Grenoble), visiting a petrol station (from 32% in Milan to 13% in Athens - only 4% in Grenoble), staying in home workshop/garage (12%/12% in Milan, 13%/9% in Helsinki, 5%/16% in Basel - only 5%/3% in Grenoble), and engaging in heavy outdoor/indoor air activity/work (34%/21% in Helsinki, 25%/19% in Basel, 10%/25% in Milan and 11%/16% in Athens - but only 5%/5% in Grenoble).

Men vs. Women:

In every city women used significantly more perfumes/deodorizers/after shaves than men. In addition to this in Athens women used more frequently kitchen extract fans. In Basel women used more frequently electric clothes dryers, and men visited more frequently home workshops, garages, petrol stations and car washes. In Helsinki women were closer to photocopiers/laser printers and used more cleaning chemicals, while men were doing more grilling, and visited garages, petrol stations and car washes more. In Milan women used more frequently glues, cleaning chemicals, while men visited home workshops, garages, petrol stations and car washes more. In Prague women used more often photocopiers/laser printers and wore more often newly dry cleaned clothes, while men were more involved with garages, car washes and heavy outdoor work.

Diary and Exposure Samples:

Significant differences in the equipment use and activities between the *Exposure* and *Diary samples* are unwanted, yet there are some. In Athens vacuum cleaners were used, and petrol stations and car washes visited more frequently within the *Diary sample*. In Basel humidifiers (and fireplaces) were used more within the *Exposure sample.* In Helsinki vacuum cleaners were used more frequently within the *Diary sample*, workplace air conditioners within the *Exposure sample*. Painting, deodorant/perfume/after shave use, staying in home workshop, visiting petrol station and heavy outdoor activity were all more frequent activities within the *Diary sample*. In Milan gas stoves were used more frequently within the *Diary sample* and vacuum cleaners within the *Exposure sample*. Visits to petrol stations, and heavy outdoor and indoor activities were also more frequent among the *Diary sample*. Also in Prague the frequency of vacuum cleaner use was higher in the *Diary sample*, gas (coal and wood) heater use in the *Exposure sample*. Painting, using of glues, deodorant/perfume/after shave, wearing newly dry cleaned clothes, grilling, and heavy indoor work were more frequent within the *Diary sample*, while staying in home workshop and garage were more frequent within the *Exposure sample*.

Durations of Equipment Use and Activities at Home and Workplace; Annex II: Tables 10./A...P

In these tables the question answered is; how much time (per 24 h) were those *Exposure* and *Diary sample* subjects, who had engaged in the equipment use or activities of concern, reported using or doing during the 48 h follow up period. Those, who had not engaged in the equipment use or activities of concern, are excluded from these tables.

Comparing the Centres:

At home the average use of gas stove ranged from 0.72 h (Grenoble) to 1 h (Milan) per day. In Athens, Basel, Grenoble and Prague the kitchen extract fan was used for the same duration as the stove. In the Extract fan use was only half of the stove use, and in Helsinki the fan was obviously used for general air exchange, nearly 8 h per day. Unvented gas water heaters were used for only short periods, 0.3...0.6 h/d, in Basel and Prague, but for 5 h/d in Grenoble and Milan. The difference probably reflects different appliance types.

The average use of supplementary room heaters was either very short term, 1-3 h/d, or the heater was on for much of the day,12-24 h/d, and was obviously replacing insufficient (or nonexistent) main heating. Gas and fuel oil heaters were the most used. Ventilating by open windows was also either shorter term, 4-7 h/d, in Athens, Grenoble, Milan and Prague, or longer term, 10 h/d in Basel and Helsinki. The wood heated sauna was heated in Helsinki in half of the time of Basel - probably due to longer cultural experience.

In the workplace the supplementary heaters were used either for a few hours only or they were replacing the main heating system (if any). If windows at the workplace were kept open, they were kept open for 1.5 (Grenoble) to 6 h/d (Helsinki). Air conditioners, humidifiers and air cleaners are also divided into two categories, those used by need and control of the occupants for a few h/d (Athens), and those integrated into central, continuously operating ventilating systems and running for most or all of the working day (Basel ... Prague).

Men vs. Women:

In Athens women, who engaged in these activities, spent in average significantly less time in grilling and home workshop than men. Also in Basel women spent less time in home workshops. In Helsinki men run the kitchen extract fans for longer times than women, and spent more time in air conditioned workplaces and garages. With the exception of Grenoble, men report to spend less time near a gas stove, but only in Milan was this difference statistically significant. In Prague only some men used home coal/wood stoves, and only some women used home air humidifiers. The differences in keeping the windows opened was rather small between the genders, except in Prague, where women kept their windows open (8 h/d) over double the time of men (3.7 h/d). Women also spent in average more time in air conditioned or humidified workplaces.

Diary and Exposure Samples:

Significant differences in the length of activities between the *Exposure* and *Diary samples* are unwanted and luckily there are only very few: In Basel heavy indoor or outdoor activities lasted longer within the *Diary* than the *Exposure sample*. In Milan, gas stove use duration was longer within the *Diary sample*.

Levels and Causes of Annoyance from Air Pollution, Annex II: Tables 11. and 12.

These tables are based on subjective evaluation of the levels and causes of air pollution annoyance experienced by the subjects in the *Exposure* and *Diary samples* during the 48 h monitoring period. The levels of annoyance are probably more based on the culture and experience than on absolute air pollution levels. This makes any comparison of these levels between the centres questionable at best. However, comparison of the annoyance caused by air pollution in the same subjects in the different microenvironments (home, work, commuting) and from different causes, should suffer much less from these problems, and therefore contain meaningful information.

These qualifications having been expressed, the lowest level of annoyance (see Annex II: Table 11.) in each microenvironment was experienced in Grenoble, followed closely by Helsinki. The highest levels of annoyance in home and commuting microenvironments were experienced in Prague, work environment in Athens. The subjects in Athens were less annoyed by air pollution in their homes than the subjects in Basel and Milan. The subjects in Basel were much less annoyed by air pollution in their workplaces than the subjects in Milan and Athens. The subjects in Milan were more annoyed by air pollution in commuting than the subjects in Athens and Basel.

The rank order of the degree of annoyance between the microenvironments was, from most to least annoying, commuting > work > home, in each city except in Prague, where work environment was considered less annoying than home environment.

The subjects were also asked for the main cause of air pollution annoyance in each of the three microenvironments. Annex II: Table 12. presents the distributions of these causes for those subjects in each city, who expressed higher than average level of annoyance in the microenvironment of concern. In the **home microenvironment** “dust” was the leading cause of higher than average air pollution

annoyance in Helsinki, Prague Grenoble, and Milan. In Basel the leading causa were “(traffic) exhaust gases” and in Athens “other”. “Chemicals” ranked lowest in each city. In the **work microenvironment** “dust” remained the single most important cause in Helsinki and Milan. In Prague and Basel “exhaust gases” were the most important causa, and in Athens and Grenoble “other”. Again, “chemicals” ranked the lowest, except in Milan, where “exhaust gases” ranked below “chemicals”. The high proportions of “other” causes indicate that for the workplace evaluation we were not able to provide the subjects an ideal selection. The most obvious and intriguing omission among the causes is ETS, and it is probably safe to assume that most of the “other” causes in homes and workplaces are indeed ETS.

While the obviously most important causes of air pollution annoyance in the **commuting microenvironment** are “exhaust gases”, it is interesting to look at the second most important. Not unexpectedly “dust” is the second most important cause in Prague, Helsinki, Grenoble and Milan, “other” (ETS?) in Athens and Basel.

5.4. Time-Microenvironment-Activity Data

The data tables presented in this chapter are based on the analyses of the 48 h time-micronevironment-activity-diaries (TMADs) that were applied to all Exposure and Diary subjects of the *EXPOLIS* study in each city. for each city the fraction of individuals entering given microenvironments or engaging in given activities

The tables show major descriptive results of the marks given in the two consecutive 24-hours-TMAD. Annex II: Tables 13./A..P present the number of subjects indicating the respective microenvironment/activity (M/A) during the 48-hours period and the distribution of total time spent in an M/A both for the whole sample (all) and for the "doers" only. "Doers" are defined as those subjects that indicate to have been engaged in the M/A at any time.

There is large variability across the centres both in the fraction of doers and the number of hours spent in M/A. The modal split for traffic shows that in Grenoble and Athens only 51% and 58% report any time spent in bike/walk, whereas on the other extreme, 93% of the Basel participants engaged in bike/walk. Furthermore, doers in Basel spent up to twice as much time in bike/walk as those from the other places. On the other hand, Basel subjects were the least likely to use the car during a 48-hour period (53%) compared to Athens (80%) and Grenoble (81%). Use of public transportation was reported by 48% in Basel, which is twice the fraction observed in Athens or Grenoble.

Among those who spent time in cars there was again a wide range in the average duration, both within and across cities. The 90th percentile was up to 15 times larger than the 10th percentile within a centre. Compared to Basel (about 37 minutes per day) the median time spent in cars among doers was 50 - 80% higher in the other places. The overall time spent in traffic was remarkably evenly distributed (1.5 - 2.0 h/d as the median, 1.9 - 2.2 h/d as the mean) between the cities. Time spent indoor at home made up the largest fraction of an average day with similar median values in Basel, Milano and Helsinki (about 13 hours per day), reaching a maximum in Athens where participants indicated more than 15 hours per day home indoors. Overall, 21 - 22 hours per day were spent indoors.

The median total time spent outdoors was highest in Basel, Athens and Grenoble with 1 hour per day with very similar distributions within city. The whole distribution of time spent outdoors was shifted towards lower values both in Helsinki and particularly in Milano (median value 35 minutes per day).

The number of diary participants reporting smoking during 48 hours varied from 0% in Grenoble (where smokers were excluded from the study) up to 31% in Milano. It should be kept in mind that these figures do not represent the prevalence of smokers in the respective populations as smokers were underselected (as described earlier) in the process of inviting participants.

Passive smoking exposure within 48 hours had been indicated by only 27% in Helsinki and 40% in Grenoble, compared to 53 - 64% in the other areas. Among the passive smoking exposed, time of exposure varied across regions with a highest median value in Milano (1 hour per day) and Athens (0.4 hours per day). The 90th percentile of passive smoking exposure at work was about 2 hours in Basel, but reaching much higher values in the other places (up to 9 hours per day). Also passive smoking prevalence may be affected by the underselection of smokers in the *Exposure samples*.

Annex II: Tables 14./A...P show the same data, stratified however by gender.

Apart from Basel, where almost everybody reported having engaged in walking/biking, women were clearly more likely to report walk/bike. In Athens the other modes of traffic did not differ by sex. In Basel, Helsinki and Milano, however, women were more likely in public transport and men more likely to report cars. In Grenoble there were no differences between the genders.

In Athens, Helsinki and Milano, male car users spent more time in the car than female users.

Men were less likely to report cooking than women in all centres and if they cook, men spend significantly less time doing so than women in all centres except Grenoble.

In all centres, women spent clearly more time indoors at home compared to men. This was particularly the case in Athens.

Annex II: Tables 15./A...P indicate the same time-microenvironment-activity data stratified for the *Exposure* and *Diary sample*.

Whereas the mode of traffic was similar across the two samples, in Basel and Athens participants of the more demanding exposure assessment were clearly less likely to walk or to take public transportation but more likely to use the car in Helsinki. In Milano the exposure sample was more likely to use the bus. In general those carrying the exposure case did walk or bike for shorter time periods than the diary sample. In all centres except Helsinki the exposure sample spent clearly 1 - 2 hours more time indoors.

Finally Annex II: Table 16 presents a comparison between all centres of the time spent in different microenvironments and activities. The most striking outcome of this comparison is, how little the differences between the centres are in terms of the average total time spent in transport, indoor at home, at work and outdoors.

General remarks:

The interpretation of these data should be done cautiously as we have not yet taken into account other cofactors which may explain the univariate differences observed (age, sex, socio-economic status, working status etc.). It should also be acknowledged that we have observed remarkable within-subject variability between day 1 and day 2, which is shown in the coefficient of variation in the table except for the time spent indoors. This variability between days within a subject was, on average, 50 - 100%. Furthermore, the obvious impact of the more demanding exposure assessment study on the time activity patterns has to be further addressed. It will be of importance in modelling/simulating population exposure distributions.

5.5. Microenvironmental and Exposure Distributions

$PM_{2.5}$, Annex II: Tables 17. and 17./G

Annex II: Table 17. presents a comparison of $PM_{2.5}$ levels and distributions between the cities and in the personal “private” and personal “workday” exposure samples with the microenvironmental samples from indoor and outdoor of the home and the workplace.

All personal and microenvironmental $PM_{2.5}$ levels are on average 50% or more lower in Helsinki than in the other cities.

Looking at the data from Basel and Helsinki one can see that the personal "private" exposure levels have similar distributions with the micro-environmental levels inside the home. In Basel the personal "private" exposure levels are about 7 $\Phi g/m^3$ higher than the microenvironmental levels inside the home, in Helsinki the two levels are in the average identical, and in Prague the personal "private" exposure levels appear to be about 5 $\Phi g/m^3$ lower than the home indoor levels.

The average and median personal "workday"levels and distributions are also similar to the microenvironmental levels in the workplace. In Basel these levels and distributions agree within about 1 $\Phi g/m^3$, in Helsinki the personal "workday" exposure levels are about 3 $\Phi g/m^3$, in Prague 5 $\Phi g/m^3$ higher than the workplace microenvironmental levels

The home indoor levels appear to follow the home outdoor levels, but at a 1/3 higher level, and the 90^{th} percentiles of the home indoor levels are usually much higher than the respective outdoor levels. The most obvious reason is smoking.

Annex II: Table 17./G shows some special features of the Grenoble $PM_{2.5}$ database. Comparing the total personal $PM_{2.5}$ exposure level (personal cumulated) to the level accumulated from indoor exposure (personal indoor) highlights the fact that the indoor exposure is absolutely dominant even for $PM_{2.5}$ from outdoor sources. Comparison of the gravimetric and reflectometric $PM_{2.5}$ data (winter) shows the similarity of the distribution but difference of the level, i.e. reflectometric data needs to be calibrated against gravimetric data.

VOCs, Annex II: Tables 18./A...P

The different VOC sampling and analysis method used in Basel compared to the other centres is reflected in the facts that 16 of the 30 target compounds (Annex II: Table 18./B)could not be analysed in Basel, but 3 additional compounds were analysed. In Annex II: Table 18./H of the Helsinki data those compounds that could be detected in fewer than 10% of the samples have been omitted, and levels below 1 $\Phi g/m^3$ have been marked as <D/L or below detection limit. 1 $\Phi g/m^3$ has been selected as the general detection limit for all compounds because it is approximately correct and the exact detection limit - which depends on the compound, and the volume and whole VOC matrix in each sample - is too complex for practical application. The same detection limits apply also to the VOC samples from Athens, Milan and Prague. No VOC samples were collected from Grenoble.

The first obvious finding from all these tables is that with very few exceptions the home outdoor levels of TVOC and each individual compound are clearly lower than the personal, home indoor or workplace levels. In Athens the workplace levels of Alkanes tend to be lower, aromatics higher and other VOCs similar to the home indoor levels. In Basel and Prague the home indoor and workplace levels are similar for all measured VOCs. In Helsinki the workplace levels are consistently lower than the home indoor levels. In Athens, Basel, Helsinki and Prague the personal exposure levels are compound by compound higher than or equal to the workplace VOC levels, and follow most closely the home indoor levels. In Athens, Basel and Prague the general rank order of the microenvironmental and personal VOC levels is personal > home indoor > workplace > outdoor. In Helsinki the personal levels are usually lower than the home indoor levels.

The approximate rank order of personal VOC exposure levels between the cities is Athens > Prague > Milan > Basel ∃ Helsinki.

CO exposure levels and distributions (undergoing analysis - to be added later)

5.6. Personal exposure determinants

5.6.1 Introduction

The main reason for investigating air pollution exposure in human populations is the fact that this exposure is known to have adverse health effects. The effects, of course, depend on the pollutant present in the ambient air and on its levels. So far, several studies have investigated health effects of air pollution. From the beginning of this century until about the end of the 60's very high outdoor pollution levels have been observed in the industrialised world (mainly in Northern Europe and North America) and several severe episodes of air pollution could have very important acute effects including sharp increases in mortality (Ware *et al.* 1981). Since then, outdoor air pollution levels have decreased. During the last decade a revival of research in air pollution health effects has been observed. It has consistently been shown that outdoor pollution levels, at current lower to moderate levels, have adverse health effects and this has especially been demonstrated for ambient particulate matter concentrations (WHO 1998). The reviewed interest led also to studies on indoor air pollutants which may be generated indoors or penetrate from outdoors (Sundell 1994).

Although results from studies investigating outdoor pollution health effects do not until now, take personal exposure determinants (other than contamination of outdoor air) and indoor pollutants into account, they still estimate the effect of ambient air pollution variability on health indicators at population level and are thus useful for public health protection. However, individual exposure is determined, to a probably greater extent, by indoor air quality, time-activity patterns (e.g. time spent commuting) and behavioural aspects (e.g. smoking or ETS exposure).

In a study where individual exposure to the pollutant studied is not taken into account, errors are introduced. These may just represent noise and lead to underestimated effects or may lead to systematic errors depending on the study design.

In this section an attempt is made to estimate the most important determinants of personal exposure to the pollutants measured in *EXPOLIS*. These include outdoor pollution levels, home and work measurements, variables characterising time-activity patterns, passive and active smoking exposure, some indoor sources etc. This investigation is limited by the small number of subjects, especially in centres other than Helsinki.

5.6.2. Methods

EXPOLIS project field work was conducted in 7 cities in Europe. The results in this chapter are preliminary and present results on three cities: Helsinki, Basel and Athens.

Regression modelling was used to address the issue of exposure determinants. Personal exposure measurements during day time and during nigh time were used alternatively as dependent variables. Because the distributions of these variables are positively skewed, they were log-transformed in all analyses.

The modelling approach has to be used because univariate associations (illustrated by two-dimensional tables) are severely confounded. There are many important variables associated with exposure, which

are also inter-related. Thus, if we want to assess the importance of the contribution of indoor air quality to personal $PM_{2.5.}$ exposure, we must take into account all other exposures of an individual. The same is true for other important exposure contributors mentioned above.

In the present analyses the following variables were used as independent variables:

For the models investigating daytime personal exposure:

1. Ambient levels of particulate matter, as measured in a representative fixed site. $PM_{2.5.}$ was not available and different methods for PM measurement are used in different cities. Thus for Helsinki PM_{10} levels were used, which must be well correlated with $PM_{2.5.}$ and in Athens Black smoke levels, which have been shown to correlate very well (r=0.84) with PM_{10}.
2. Work and home location. In the questionnaire the home and work location were described as: suburban in a suburb with low-rise homes; suburban in a suburb with high-rise homes; industrial areas, downtown urban area. These categories were considered as having an ascending order of air pollution level (from 1 to 4) and home and work location were entered in the model as ordinal variables. Because not all subjects were working, a dummy variable indicating work status (yes/no) was also included.
3. Traffic near home and work place. In the questionnaire a question recorded the traffic density in the nearest street to the home and work in 3 categories: light, moderate, heavy. These categories are considered as reflecting traffic generated pollution exposure ordered from smaller to heaviest (1-3) and were also entered as 2 ordinal variables in the model.
4. Smoking exposure. Two sets of variables were introduced in the model alternatively: One set included smoking at home (yes/no) and smoking at work (yes/no). These variables represent exposure to both active and passive smoking which takes place indoors at home or at work. The second set was constructed from the time-activity diary and it represented length of time of active and passive smoking exposure, ordered in tertiles of the distribution (from 1 to 3).
5. Indoor home and work measurements. The measurements done within the *EXPOLIS* framework for home indoors and workplace were used.

5.6.3. Results

Athens

In Annex II: Table 19.-1/A we may see that night-time personal exposure to $PM_{2.5}$ and home indoor measurements are increasing with increasing traffic near the home. Personal daytime is not so much associated with traffic which may be expected and unexpectedly there seems to be no association with home outdoor measurements. This needs to be further investigated.

In Annex II: Table 19.-2/A similar results are observed for personal night-time exposures with home location. In Annex II: Table 19.-3/A, both personal daytime and workplace measurements are increasing with increasing traffic density near the workplace but the associations are less consistent with "work location". In Annex II: Table 19.-4/A we may see that univariate associations of community habits based on time-activity diaries and personal $PM_{2.5}$ exposure are not clear. In Annex II: Table 19.-5/A, we can see that smoking at home and at work affects both personal and indoor measurements. Also, smokers are more exposed to $PM_{2.5}$ than non-smokers but no dose-response is observed with ETS exposure. In Annex II: Table 19.-6/A we may see that the existence of gas appliances and air conditioning seem to increase PM exposure, but in Annex II: Table 19.-7/A it is seen that there is no clear association of ambient BS levels and personal exposure.

In Annex II: Table 19.-8/A the results of multiple linear regression models for day-time personal $PM_{2.5.}$ exposures are shown. Although the variability explained in the log-transformed dependent variable is not small, especially in the first model (34%), there are no statistically significant determinants of exposure. Furthermore, the coefficients seem rather sensitive to the variables included in the model, which may be a result of the small number of exposure determinants.

In Annex II: Table 19.-9/A the multiple linear regression results for night-time personal exposures are shown. These results are more robust, probably because there are less exposure determinants for night-

time exposure. Thus, the second model explains 55% of the personal exposure to $PM_{2.5.}$ and home location and ETS exposures are positively associated with personal exposure to ETS although only home location to a nominally statistically significant degree.

Basel

In Annex II: Tables 19.-1/B-2/B we can observe that traffic density near the home is associated with personal (daytime and nighttime) exposure but not with home indoor and outdoor environmental levels, while home location is not monotonically associated with exposure measurements. In Annex II: Table 19.-3/B we see that heavy traffic density near work is related to higher personal and workplace exposure, but work location does not seem to be consistently associated with exposure. From Annex II: Table 19.-4/B it may be seen that time spent in different means of transportation for commuting to work does not seem to be monotonically associated to personal $PM_{2.5}$ exposure. In Annex II: Table 19.-5/B it can be seen that smoking at home or at work, active smoking and prolonged ETS exposure are all associated with significantly higher personal and indoor $PM_{2.5}$ levels, while from Annex II: Table 19.-6/B we see that the existence of gas appliances and air conditioning do not alter the exposure level, whereas having the windows open for longer time is slightly associated with higher exposures. From Annex II: Table 19.-7/B it can be seen that ambient levels of PM_{10} are not consistently associated with personal exposures.

Annex II: Table 19.-8/B shows the results of multiple regression models for day-time personal exposure. The models explain 64% of the variability in personal exposure. Environmental level of $PM_{2.5}$ at the workplace is the most statistically significant exposure determinant in both models ($P<10^{-3}$), followed by ambient PM_{10} measurement at a fixed site (P=0.06). Annex II: Table 19.-9/B shows the corresponding results for night-time exposure. The explained variability is 51% in the second model. Home indoor measurements is the most important determinant of exposure (P=0.001), followed by home location (P=0.16).

Helsinki

Annex II: Tables 19.-1/H to 6/H show the levels of personal and - in some instances, where it is relevant - environmental measurements of PM2.5., by the categories of possible exposure determinants. Several descriptive statistics are shown: the number of subjects in each category, the mean, standard deviation and the median. Statistical evaluation is not done in these Tables because, as mentioned in the Methods section, these associations illustrated here are heavily confounded. However, a few comments describing the data are given.

In Annex II: Table 19.-1/H we see that traffic density (heavy) near home seems to be associated with increased personal night-time $PM_{2.5.}$ exposure, and home indoor levels. From Annex II: Table 19.-2/H it may be seen that downtown residences are associated with relatively high average home indoor and personal nightime exposures. These, however, appear to be caused only marginally by outdoor air, and probably mostly by indoor or personal sources. In Annex II: Table 19.-3/H it can be seen that traffic density near work does not seem to be clearly associated with PM exposure, while data for work location were not available for Helsinki. Annex II: Table 19.-4/H shows that length of commuting by different means of transportation is not clearly associated with personal $PM_{2.5.}$ exposure. In Annex II: Table 19.-5/H we may see that smoking at home affects both personal and home indoor $PM_{2.5.}$ levels and the same is true for smoking at work. Active smoking affects personal exposure but not home levels (since presumably smokers smoke outside). Time exposed to ETS is associated with personal exposure as well as home indoor levels of $PM_{2.5.}$ In Annex II: Table 19.-6/H we can see that the existence of gas appliances is not associated with a different level of $PM_{2.5.}$ and also no striking differences are noted for the existence of air conditioning and duration of opening windows. In Annex II: Table 19.-7/H we observe a consistent increase in the median levels of personal exposure with increasing quintiles of ambient PM_{10} levels.

In Annex II: Table 19.-8/H we see the results of multiple linear regression models for day-time personal exposure. Both modes explain around 45% of the variability in personal exposure and the coefficients are rather insensitive to model specification. The levels of $PM_{2.5.}$ in the indoor home and work environment are very statistically significant determinants of day-time personal $PM_{2.5.}$ exposure. Smoking at work and total exposure to active smoking are also statistically significant exposure determinants. PM_{10} levels in the outdoor air and passive smoking exposure are important determinants (especially in the second model) but do not reach the nominal level of significance.

In Annex II: Table 19.-9/H the results from regression models for night-time exposure are shown. The second model explains 64% of the personal night time $PM_{2.5}$ exposure. The outdoor PM_{10} levels, home location, home indoor levels and length of exposure to active smoking are all statistically significant determinants of exposure.

From the discussion on air pollution and health research some gaps and future needs were identified:

1. Two air pollutants, SO2 and CO have been almost forgotten. New evidence suggests that we need more information about their exposures and health effects.
2. There is a strong need for assessing long-term heath effects. A cohort study is the most straightforward design, but an expensive and long duration undertaking. It should be carefully designed and planned in detail on one hand to maximise the benefits of the wide qualitative and quantitative variety of air pollution across Europe, and on the other to control the formidable socioeconomic, cultural and climatic confounders. Imaginative and novel research approaches should be considered.
3. Better understanding is needed on the association of personal exposure with ambient air pollution and other indoor, outdoor, personal and socioeconomic factors.
4. Air pollution exposure modelling techniques should be developed and evaluated for the exposure assessment needs of air pollution epidemiology, and risk and environmental impact assessments.
5. Better understanding is needed on the association of biomarkers of exposure with specific air pollution exposures, other exogenous exposure pathways and endogenous exposures.
6. European research on the mechanisms of the observed health effects of exposures to the present levels of air pollutants should be strengthened.
7. The link between research results and the needs of policy formulation, standard setting and emission control should be solidified. Communication and feedback between the policy makers and researchers should be increased.

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5.7. Exposure Simulation

5.7.1. Introduction

This section describes the development of a model system for the assessment of integrated (sub-)population exposure distributions for selected air pollutants, based on Monte Carlo simulation techniques. The model system is intended for use in the *EXPOLIS* study and has been developed with the goals and collected data of *EXPOLIS* in mind. However, the resulting modeling framework is flexible enough for use in other studies and applications.

It is our expectation that the described exposure model will be useful in scenario studies and in assessing the public health gain of environmental policy options (in terms of population exposure and/or population attributive risk). However, real validation of the model has not yet taken place and the model should therefore be seen as a starting point for the modeling exercises that will take place. The *EXPOLIS* databases form an excellent base for further refinement and extension of the model.

Section 5.7.2 describes the developments in the field of exposure modeling over the last decades. It also gives an overview of the different types of exposure models and briefly introduces the concepts behind these models. In *section 5.7.3* a description of the modeling concept that is chosen within the *EXPOLIS* study is presented together with an explanation of the Monte Carlo simulation method. Finally, *section 5.7.4* introduces the modeling framework that has been build within the *EXPOLIS* study and a stepwise approach for the user to build his own exposure model in the presented framework.

5.7.2. Short Overview of Human Exposure Models

Aspects of Human Exposure Modeling

Human exposure models have proven to be a valuable and cost-effective tool for assessing potential exposures experienced by individuals, both for research and regulatory purposes. A model is best viewed as a tool for the investigator. It is an aid in understanding exposures and the factors that

influence them (Ryan, 1991). The place of exposure models in the chain from emission to effect is presented in the general risk model, Figure 5.7.-1 (Van Scheindelen et al., 1995; Ott, 1985; Lioy, 1990):

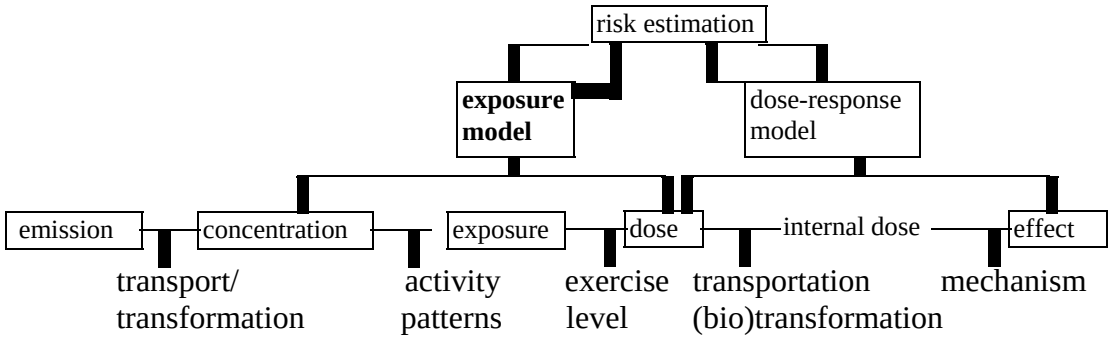


Figure 5.7.-1 General environmental health risk model.

Most important components of this scheme for exposure modelling are concentration, exposure and dose.

Concentration (the amount of material contained in a fixed volume, expressed in mass per volume (Ryan, 1992)) is a clearly defined parameter, but exposure and dose are terms that are not defined in a very consistent way. Duan, Dobbs and Ott proposed definitions that would be widely applicable (Duan et al., 1989). Before coming to a definition of exposure and dose they define a number of parameters which determine the specific form of exposure and dose:

1. *the target* (who): any physical entity occupying space;
2. *the pollutant* (what): specification is determined by the observation used to take the sample and is limited by the availability of technologies;
3. *the medium* (how): the pollutant might be carried in several different media. There may be different penetration properties and health effects; multimedia exposure and the health effects may be more relevant than single-medium exposure;
4. *the route* (how): the same pollutant carried by the same medium can impact the target via different routes; the penetration properties and health effects might be different;
5. *the contact surface* (where): the part of the external surface of the target where the penetration can occur, with homogeneous pollutant concentration. For air pollutants generally the respiratory tract epithelium is of interest.
6. and *the time frame* (when): The averaging time may be relevant for potential health effects.

Exposure and dose can now be defined:

Exposure *is the contact between a target and a pollutant on a contact surface.*

Exposure requires the simultaneous presence of a concentration of pollutant and a target receptor (Ryan, 1991).

Dose *is the penetration of the pollutant in or out the target via the contact surface.*

It is a function of the concentration of the pollutant in the environment, the presence of an individual in that location, and the physiological state of the individual.

Different exposure and dose measures can be derived with different time frames. For most purposes, knowledge of dose is more relevant than knowledge of exposure, but usually it is more difficult to measure dose than to measure exposure. Therefore, exposure is very often assessed as a proxy for dose.

The appropriate choice among the variety of possible definitions of exposure and dose depends on the specific goal of the study, the implication for dose assessment and the health effects of interest. If the dose-response relationship suggests that the health effect is cumulative and linear, the average exposure

or the integrated exposure should be considered. If the health effect is non-linear, those exposures might be inappropriate.

Also, the feasibility of measuring the exposure is important. If the best instrument available can only record 24 hour integrated exposure, the maximum one hour average exposure probably cannot be assessed (Duan et al., 1989). However, with knowledge of the stochastic structure of the concentrations it might be possible to measure exposures with a longer averaging time and use such data to impute shorter-term averaged exposures (Ryan, 1991).

A complete description of exposure requires knowledge of the *magnitude* of pollutant concentration in the exposure environment, *duration* of exposure, and the *time pattern* of the exposure (Ryan, 1992). The record of an individual's exposure throughout the day is called an *exposure profile* (Ott, 1995). For information on health effects the distribution of individual exposures is of interest, but to protect a certain population from exposure, the exposure distribution for a population is more important (Ryan, 1992, Ott, 1985). Human exposure models can be used to assess these distributions.

Human Exposure Models

Ryan described three classes of models that might be used to model human exposure to air pollutants: *statistical models, physical models, and physical-stochastic models*.

For human exposure modelling the *statistical approach* requires the collection of data on both actual exposures and factors that possibly influence the exposures. Sufficient variability is necessary to develop a useful predictive model. Bivariate and multivariate correlation's between and among the measured variables can help in selecting factors; collinearity problems might appear and must be avoided. Model building can be performed with variables that contribute sufficient variability to the total. Modellers must be careful with extrapolation outside the database because of not having evidence that the model is applicable in other situations.

The *physical approach* to modelling human exposure to airborne pollutants is very different from the statistical approach, because the researcher defines the physical model for the system *a priori*. After defining the model it is transformed into an abstract mathematical form. This approach is a qualitative improvement over the statistical approach because of the use of concrete physical ideas. Still there are unknown components of total exposure in these models. There is no prediction of uncertainty.

Examples of physical models of human exposure are the following. Exposure experienced by an individual is related to the outdoor concentration of the pollutant in question. Total exposure can then be a linear function of the ambient concentration: E=aC+b. With personal and ambient field measurements the parameters a en b can be determined. Sexton et al. (1983), Ryan et al. (1983) and others later draw relationships between a and b and physical variables (air exchange rate, presence of sources independent from the ambient pollution level etc.).

More complex is the *microenvironmental approach*, in which total exposure is a sum of the exposures within various microenvironments. One example of this is NEM (Johnson, 1995). Letz et al. (1984) added information about physical parameters influencing microenvironmental concentrations of some pollutants.

The *physical-stochastic approach* is a relatively new type of modelling. Exposure distributions can be computed combining deterministic parts of the physical models with the uncertainty of a finite model of physical processes and human activities. It tries to account for the probabilistic or stochastic nature of the physical aspects of human exposure. Essential components of the total exposure can be accounted for and information on unseen components can be gathered. Human behaviour and air pollution are probabilistic processes. For (sub)populations behaviour can be predicted with good accuracy and

precision, but for the actions of a particular individual it is much more difficult. Examples are SHAPE (Ott et al., 1984) and SIMSYS (Ryan and Letz, 1991).

5.7.3. EXPOLIS Exposure Model

The choice for the modeling framework that is used within the *EXPOLIS* study has been guided by a number of considerations. The most important was the availability of information resulting from the *EXPOLIS* study. Furthermore, different research centers in Europe should be able to use the model for calculations on distinct (sub-)populations thus requiring the model to be flexible both in required data input and in data output. The time constraints given by the duration of the project played a role in the possible complexity of the model.

A summary produced by Ryan (1991) is presented in Table 5.7.-1.

Table 5.7.-1. Basic properties of the three general exposure model types.

Parameter	**Model Type**		
	Statistical	**Physical**	**Physical-Stochastic**
Method of Formulation	Descriptive analyses; Hypothesis testing	Physical laws	Physical laws and statistics
Required input	Collected Data on human exposure	Knowledge of important parameters and their values in system to be modelled	Knowledge of important parameters and their distributions in the systems to be modelled
Advantages	Makes use of real data in the model-building process	True model developed from a priori considerations	Model developed from a priori considerations; stochastic feature allows uncertainty to contribute, reducing importance of researcher biases
Disadvantages	Requires data on hand for model building; extrapolation beyond data base is difficult	Includes researchers biases; must be validated	Requires much knowledge of the system; must be validated.

Examples	Various epidemiological studies, such as the Harvard Air Pollution/Lung Health Study Stepwise regression models which include physical parameters in the regression model	National Exposure Model (NEM) Indoor Air Quality Mass Balance Models	SHAPE SIMSYS pNEM

Given these considerations and based on a literature review and modeling experience available at RIVM a choice has been made for a basic microenvironmental model using a physical-stochastic approach. The microenvironment model we use was first described by Duan in 1981, and further developed by Ryan, Spengler and Letz (Duan, 1981, Ryan et al., 1986). Further work using this approach has been carried out at RIVM in a project called MOSES (Microenvironmental Oriented Simulation of ExposureS) (Melse et al., 1996).

Model Description

In our microenvironmental model the personal exposure is the total of partial exposures determined by the combination of the concentration of the air pollutant in a microenvironment and the time an individual spends in that microenvironment. In each microenvironment a homogeneous air pollutant concentration is assumed.

The notation in the model is as follows:

$$E_{tot} = \sum_i f_i * C_i \qquad (1)$$

where: E_{tot} = total personal exposure, expressed as the average concentration for the integration period

f_i = time fraction spent in *i*-th microenvironment

C_i = concentration in *i*-th microenvironment

For indoor microenvironments the concentration distribution *without* attribution of sources is required. In addition to that information on the concentration distribution of each relevant indoor source is needed.

In case no direct information about the concentration in an indoor microenvironment is available, the indoor concentration, C_i, can be estimated by:

$$C_i = C_{out} * p_i + S_n \qquad (2)$$

where: C_i = concentration in *i*-th microenvironment (without sources)

C_{out} =outdoor concentration (at the front of the dwelling)

p_i = penetration of the pollutant in the *i*-th microenvironment

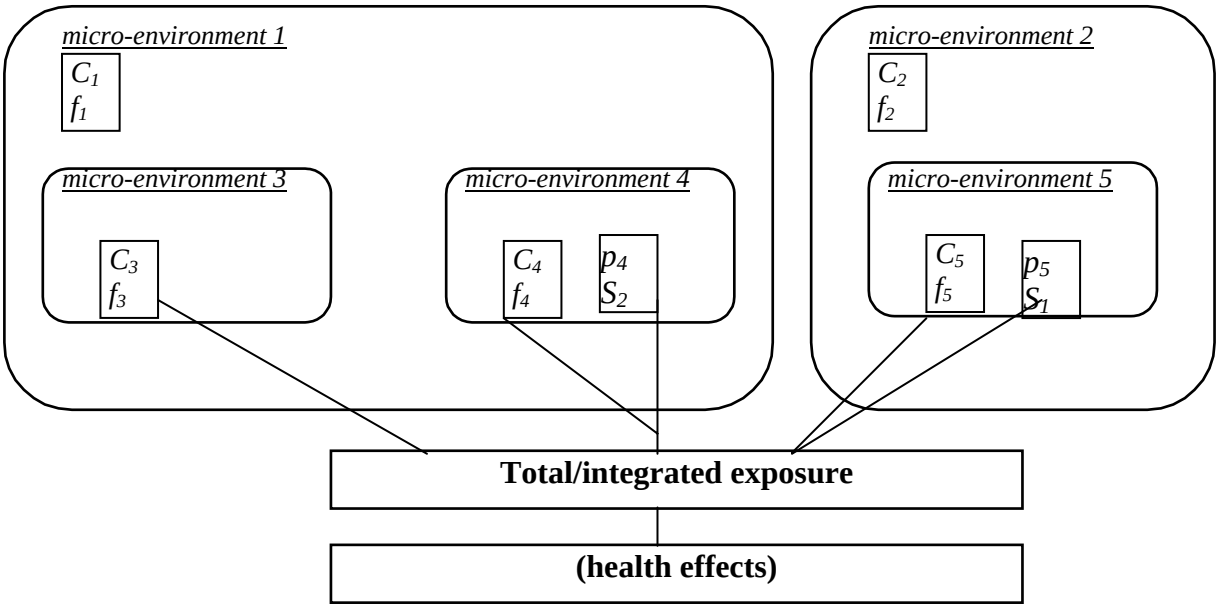
S_n = attribution of sources in the indoor microenvironment

This conceptual model and the corresponding parameters are represented in Figure 5.7.-2, where microenvironment 1 and 2 represent outdoor microenvironments. Both microenvironments require information about the concentration distribution and the distribution of the time fraction. Microenvironments 3, 4 and 5 are located indoors. The concentration distribution in microenvironment 3 is known, thus requiring the same parameters as 1 and 2. For 4 and 5 no direct information about the concentration distribution is available. An estimation of the concentration distribution can be made based on the concentration distribution of the outdoor microenvironment they are located in, the penetration of outdoor pollution into the microenvironment and, if present, additional local sources in the microenvironment.

Monte Carlo Simulation

A Monte Carlo simulation approach is chosen for the calculation of the model. Monte Carlo refers to the traditional technique for using random or pseudo-random numbers to sample from a probability distribution. With enough iterations, Monte Carlo sampling 'recreates' the input distributions through sampling. In our model a more sophisticated sampling technique is used, called Latin Hypercube sampling. In this method the input probability distributions are stratified, dividing the cumulative curve into equal intervals. A sample is taken randomly from each interval of the input distribution. An advantage compared to regular Monte Carlo sampling is that fewer samples have to be taken to recreate a valid input distribution and correlation's between distributions can be taken into account (@Risk manual, 1997).

Figure 5.7.-2. Conceptual model.



A certain type of probability distribution function is assumed for each parameter presented in the previous section. From the microenvironmental concentration distributions and the time activity distributions, random values are taken using the Latin Hypercube method.

After having sampled all relevant parameters, these parameters are combined to result in a partial exposure for a microenvironment. By summing all these partial exposures and repeating the procedure a

large number of times the integrated or total exposure distribution is calculated for a (sub)population. From this exposure measure other exposure measures can be derived, e.g. average exposure of a population plus its standard deviation, or the percentage of people exposed above a certain exposure limit (e.g. NOEL).

It is important to note that the distributions of the input parameters and the total exposure distribution represent a population exposure distribution, thus making it impossible to recreate the personal exposure of an individual.

Input Parameters

The following minimum input information is needed in our microenvironmental model:

1. definition of the micronvironments under study;
2. concentration distribution in each microenvironment;
3. time activity patterns.

Microenvironments under Study:

Which microenvironments are identified in the model depends on choices made by the researcher. Aspects that have to be taken into account when choosing the relevant microenvironments are: the application of the model, data availability, correlation between microenvironments (e.g. urban/suburban/rural) and the critical components of each microenvironment (Concentration or time or C*t).

Concentration Distribution in Each Microenvironment*:*

An assumption made in the model is that microenvironmental concentrations are distributed lognormally. In microenvironments with measured data, the description of concentration distributions requires the geometric mean (GM) and geometric standard deviation (GSD) of the microenvironmental concentration, plus the same parameters for the relevant indoor sources. The concentration distribution will be bounded by zero from below, but will have no upper limit.

In *EXPOLIS* microenvironmental concentration data is collected for:

home indoor, home outdoor, work indoor, and some information of *other indoor* and some information on modes of transport (commuting).

For the relevant indoor microenvironments where no direct information on the concentration distribution is available, an indirect estimate can be made (formula 2) using information about the outdoor concentration at the front of the dwelling (C_{out}), the penetration factor (p) and the attribution of sources in the indoor microenvironment (S_n). In *EXPOLIS*, this information can possibly be derived from questionnaire information.

The penetration factor accounts for infiltration and sinks of the pollutant, and can have a value between 0 and 1, where 0 is no penetration, and 1 is full penetration.

Shape of the distributions of sources will depend on the source, but will again be bounded from below by zero, and will have no upper limit. In the current model, lognormal distributions are assumed.

Correlations: Concentration distributions between microenvironments as well as time spent in the different microenvironments can be correlated. For example, urban and rural concentrations of $PM_{2.5}$ often have a positive correlation. In the model, a correlation matrix gives the user the possibility to specify correlations between microenvironments.

Time Activity Patterns

How people spent their time depends on several characteristics, e.g. on employment status, on age, and probably also on socio-cultural aspects. Therefore it is important to identify *subpopulations* or *cohorts*: a group of people with similar time activity patterns. In *EXPOLIS* only information is gathered on the working adult people living in European cities. Subgroups in the *EXPOLIS* study population still have to be defined.

In the model, the fraction of time that is spent in each microenvironment is described with a beta distribution.

Monte Carlo techniques using @Risk: The model is implemented in @Risk, a Risk Analysis add-in program for Microsoft Excel. Apart from Monte Carlo simulation techniques, the program is also able to perform sensitivity analyses and uncertainty analyses, in order to evaluate the robustness of the model with respect to potential changes in inputs and parameters and to provide quantitative estimates of uncertainty in the model predictions. With this software, it is also possible to build in correlations between distributions.

Assumptions: In the model assumptions are made that the air pollution concentrations and the concentration of local sources follow a lognormal distribution. The time fractions spent in each microenvironment and the penetration factor are described using a beta distribution. These assumptions are based on the literature (Ryan et. al., 1986). Whether or not the selected distributions are the best option is debatable and depends on the population under study and the microenvironments that have been selected.

For the time fractions we first checked our assumptions using the Dutch Intomart activity pattern survey (Intomart, 1995). In this survey, respondents filled a questionnaire consisting of a 24-hour diary (15 minutes resolution) and a list of questions on the person and his/her household. In the diary the respondent keeps track of the microenvironment in which he resides (7 categories), his activity (20 categories), and the level of exercise (5 categories) of his activity. The resulting database contains information on 5060 respondents that form a representative sample of the general population (Intomart, 1995; Freijer et. al., 1997). See also Annex II/B.

From the 5060 respondents we selected the respondents that resemble the *EXPOLIS* customers as closely as possible. Only working respondents in the age between 25-55 years were included in the analysis (= 1964 respondents).

The Bestfit (Bestfit, 1997) software was used for probability distribution fitting. For a given input distribution Bestfit looks for the parameters of the function that optimise the goodness-of-fit, a measurement of the probability that the input data was produced by the given distribution. Bestfit is able to calculate the goodness-of-fit statistic for more than 30 different distribution types. The distribution types were then rank ordered according to several statistical tests.

Bestfit selected the most probable distribution type for all 7 microenvironments that are present in the Intomart database. More detailed information can be found in Annex II/C. The results show that the Beta distribution is the optimum choice in only 2 of the 7 microenvironments. For one microenvironment (Indoor, not at home) it was hardly possible to select a suitable type of distribution because the input seems to be bi- or even trimodal.

This preliminary analysis shows that it is very important to think about the distribution type that you will be using in the model and the population that you want to study.

5.7.4. User Manual-Model Implementation

In this section the installation of the application and the process of building your own exposure model is described using an example model for $PM_{2.5}$ in Helsinki. The values of the parameters in this example model are taken from *EXPOLIS*, literature or experts judgement (Sexton et al., 1984; Spengler, 1985; Morandi, 1988; Quackenboss, 1989; Li, 1994). The application is not a standalone program, but is implemented in MS-Excel and @Risk. The user should therefore have a good knowledge of Excel and understand the basics of @Risk.

Installation and System Requirements

The application consists of two Excel files called '*EXPOLIS*.xls' and 'example.xls' and is issued on one floppy disc. The file '*EXPOLIS*.xls' contains the empty modeling framework while the file 'example.xls' contains the example model that is presented in the following paragraphs. The following minimum system requirements are recommended for use of the application:

- A 486 or Pentium PC.
- MS-Windows 3.1 or higher.
- 8 MB RAM and 16 MB available memory (actual and virtual). Running the model with this minimum amount of memory is quite slow and more installed memory is therefore recommended.
- MS-Excel version 5 or higher.
- @Risk version 3.5 or higher. @Risk should be installed as a 16-bit program for Excel 5.0 and as a 32-bit program for Excel 7.0 or higher.

The application can be run by starting @Risk and opening the file '*EXPOLIS*.xls'.

The application contains three worksheets and a macro. The worksheets are:

input definition of microenvironments and entry of relevant parameters for the description of each microenvironment

correlation correlation matrix for incorporating correlations between the stochastic parameters in the model

calculation calculation of population exposure distribution based on the two previous worksheets

The function of each worksheet and the requested input on each worksheet will be explained in the following paragraphs (see also Annex II/A).

Building Your Own Model

Before you start building your own model you should have a clear picture of the population that you would like to study, the microenvironments of interest and the data that you have available about each microenvironment. The following steps can be distinguished while building your model:

1. Definition of (sub)population under study and definition of microenvironments (µEs). Divide the µEs in two groups: µEs with data available about the concentration distribution and µEs that have no readily available data about the concentration distribution (should be estimated)
2. Enter parameters of µEs with known concentration distribution
3. Enter parameters of µEs with unknown concentration distribution

4. Define correlation structure in the model
5. Specify @Risk settings
6. @Risk output

Defining the (sub)Population and Microenvironments

Before you can start building an exposure model careful thought should be given to the population that you want to study and the microenvironments (µEs) that this population can move through. The most accurate results will be obtained when studying a population that is homogeneous, for example a population of office workers during weekdays. Office workers will follow more or less the same pattern during their working days. This makes it easier to define the µEs that they can move through and also reduces the number of µEs that is necessary to describe the pattern of activities of your population.

Building a model for, for example, the general population would cause more trouble in defining all µEs. Furthermore, the distributions used in the model will hardly ever be able to describe your population because the pattern of activity in a µE can differ widely between individuals in the population causing the distributions to be bimodal or take some other indescribable shape.

The choice of the population under study and the number of µEs should also be guided by the available information. The model should always contain at least one outdoor µE. This is necessary because estimation of concentration distributions in µEs is based on the outdoor concentration distribution. Try to group the µEs that most of the population visits during very short periods of time only (for example, travel by bus, train and car). Without grouping you have to gather large amounts of data while the influence of these µEs on the resulting output concentration distributions is rather small.

During the construction of your model you should always be aware that the uncertainty in subjective estimates (the input variables) has two components: the inherent uncertainty of the variable itself and the uncertainty arising from the expert's lack of knowledge of the variable. In a risk analysis model, these uncertainties are not distinguished and the combined uncertainty is an input to the model (Vose, 1996).

Divide the µEs into two groups. The first group consisting of the µEs with available information about the concentration distribution in the µE. And the second group consisting of the µEs where the concentration distribution needs to be estimated based on the outdoor concentration, the penetration factor and possible indoor sources.

Microenvironments with Known Concentration Distributions

Parameters describing the µEs with a known concentration distribution can be entered on the 'input' worksheet. This worksheet is divided into three different sections, the yellow cells can be used for entry of parameters. The upper section of this worksheet is presented in Table 5.7.-2.

In the first column the names of the µEs should be entered. In our example, three µEs have been defined: home indoors, home outdoors and work indoors. A maximum of 12 µEs with known concentration distribution can be defined.

For each µE the geometric mean (GM)(in $\mu g/m^3$) and the geometric standard deviation (GSD) of the concentration distribution have to be entered in columns 2 and 3. In the model the assumption is made that all concentration distributions follow a lognormal distribution. The lognormal distribution is bounded from below by 0.

The fraction of time that the population spends in each µE has to be entered in columns 4 and 5. In the model the assumption is made that the fraction of time follows a beta distribution, requiring a mean and standard deviation (SD). 0 and 1 bound the beta distribution. Therefore, the mean fraction of time is expressed as a percentage of the total time. The total time in the model is always 1.

Next you enter the fraction of the population who are exposed to a certain indoor source (e.g. smoking) present in the associated indoor microenvironment.

Table 5.7.-2 µE parameters with a known concentration entered on the 'input' worksheet

Micro environments with known concentration distribution										
µE		Concentration (C_i)		Fractional time (f_i)		Possible indoor sources				
		GM	GSD	Mean	SD	1	2	3	4	5
1	Home indoors	8,2	4,5	0,56	0,13	0,17				
2	Home outdoors	7,8	1,8	0,02	0,04					
3	Work indoors	7.8	2,8	0,24	0,13	0,14				
4										
5										
6										
7										
8										
9										
10										
11										
12										

Microenvironments with Unknown Concentration Distributions

If you have no direct information available about the concentration distribution in one or more µE's, an estimate has to be made. Parameters required for making these estimates should be entered in sections 2 (Table 5.7.-3.) and 3 (Table 5.7.-4.) on the 'input' worksheet.

In the first column the names of the µEs can be entered. In the model it is possible to make an estimate of the concentration distribution for a maximum of eight µEs. In our example we have defined eight µEs in this section (see above).

Columns 2 and 3 ask for the mean and standard deviation of the fraction of time that the population spends in each µE, again to estimate a beta distribution. At the end of column 2 there is an additional cell that gives the sum of the fraction of times of all µEs in the model. This cell can be used as a check for the user to see if the means of all fractions of time add up to about 1. If the total time is not exactly one, an adjustment is made to make the total of fractions one (see Annex II/C for a description of the sampling process of the time fractions).

Column 4 and 5 ask for the mean and standard deviation of the penetration factor (or Input/Output ratio). In the model the penetration factor also follows a beta distribution[1]. In this model you cannot enter 1 for the mean and 0 for the standard deviation of this distribution, but instead a value that

[1] Please note that the beta distribution is described by the parameters α_1 and α_2. These parameters are calculated using the mean and the standard deviation. The formula's for calculation in @Risk prohibit the use of the value 1, resulting in errors in the model. Instead, use the value 0.999 as an approximation.

approaches 1 and 0 have to be taken to get an outcome from the calculations. This is due to the underlying equitations.

In column 6 the user is asked to specify the outdoor concentration distribution (C_{out}) on which the estimate of the concentration distribution is based. A number of a µE from the first section of the input sheet should be entered in this column. In the example the estimates of both home and indoor (other) are based on the outdoors µE in section 1.

Table 5.7.-3. Section 2 of the input worksheet (see text)

Micro environments with unknown concentration distribution											
µE		Fractional time (fi)		Penetration factor (P_i)		Depending on	Possible indoor sources (S_n)				
		Mean	Std	Mean	Std	outdoor µE	1	2	3	4	5
13	work outdoors	0,01	0,04	1	0	2					
14	other indoors	0,05	0,06	0,06	0,2	2	0,1				
15	other outdoors	0,01	0,03	1	0	2					
16	walk/bike	0,03	0,03	1	0	2					
17	Motorbike/ Scooter	0	0,01	1	0	2					
18	car/taxi	0,04	0,05	0,8	0,3	2					
19	bus/tram	0,01	0,02	0,75	0,25	2					
20	train/metro	0	0,01	0,6	0,2	2					
	Sum of fract. times:	0,97									

The last five columns give the user the possibility to add the contribution of possible indoor sources (S_n) to the estimate of the exposure distribution. In case no indoor sources are present, these columns can be left empty.

Table 5.7.-4. Section 3 of the input worksheet (see text)

Indoor sources			
Source		Concentration (S_n)	
		GM	GSD
1	Smoking	12,01	5,74
2			
3			
4			

In this section local indoor sources can be defined, with a maximum of 5 sources. In our example we defined smoking as an indoor source. The concentration distribution of all indoor sources is assumed to follow a lognormal distribution, given by the mean (in $\mu g/m^3$) and the standard deviation.

Once the indoor source have been defined you should go back to the last 5 columns of section 2 to link the sources to their respective µE's. The default value in these columns is 0, meaning that there is no link between an indoor source and a µE. A link can be made by entering a value between 0 and 1. The value 1 means that the whole population in that µE is exposed to the indoor source. A value between 0 and 1 means that a certain percentage of the population is exposed to the indoor source. In our example, we have connected the indoor source 'smoking' to the µE 'home' by the value 0.4. This means that 40 percent of the population is exposed to smoking in the home.

Correlation Matrix

When you are building your model and/or looking at your data you might have found that there are dependency relations in your database. These relationships can be accounted for in your model using the correlation matrix in the 'correlation' worksheet.

In the matrix you can define correlations between all stochastic input variables in your model. The names of the µEs in your model are automatically repeated in the matrix.

The value of the correlation coefficients can vary between –1 and 1, where 1 means perfect positive correlation, 0 means no correlation, and –1 means perfect negative correlation. The correlation matrix forms the basis for the rank correlated sampling of your input variables during simulation.

In the example only correlation coefficients larger than (-)0.25 have been entered.

Specify @Risk Settings

A number of default settings are defined in the @Risk model. It is good practice to check the settings before each simulation that you run. The settings can be changed according to the user's wishes.

Click the 'Change @Risk settings' button on the @Risk toolbar to enter the simulation settings menu. The default settings under each tab are as follows (more information can be found in the @Risk manual):

Iterations tab:

- *# iterations* = 2000
- *# simulations* = 1

Sampling tab

- *Sampling type* = Latin hypercube

  It is important to use Latin Hypercube sampling when you have defined dependency relationships in the correlation matrix. It is not possible to define correlation's using Monte Carlo simulation. The user is referred to the @Risk manual for an explanation.

- *Standard recalc* = Expected value

- *Random number generator seed* = 1

Setting the seed to a value other than 0 has certain advantages. Providing the model is not changed, the same simulation results can be exactly repeated. More importantly, one or more distributions can be changed within the model and a second simulation run to look at the effect these changes have on the model's output. It is then certain that any observed change in the result is due to changes in the model and not a result of the randomness of the sampling (Vose, 1996).

- *Collect distribution samples* = checked

Convergence tab

- *Monitor* = checked

- *Check every* 100 *iterations*

Macro tab

- *Execute macro ?* = checked

- *Macro name:* = dcout

- *Macro executed when ?* = Before simulation

The macro 'dcout' is necessary in the model to connect the estimated concentration distribution of the indoor µEs with the outdoor concentration distribution (C_{out}) and should be run before the simulation starts.

External tab

- *Process model in:* = spreadsheet

Once the simulation settings have been defined, you should check if the output variable is correctly defined. Do this by clicking the 'Inputs by outputs' button on the @Risk toolbar. The 'Inputs by outputs table' of @Risk appears showing the input and output distributions of the model. In the output section on the left of your screen the cell S33 should be present. If this is not the case, go to the 'calculation' worksheet and activate cell S33 by clicking it. Then press the button 'Add the selected cells as @Risk outputs'.

Press the 'Run simulation' button on the @Risk toolbar to run the model.

5.7.5. Output of the Model

After running the model the software automatically opens the results window of @Risk. Here an overview of the simulation results is given. The simulation results are available as statistics, data and in graphical format. Below an example graph is given of the graphical output (Figure 5.7.-3.) generated by @Risk. The graph shows the 24-hour exposure distribution that has been simulated based on the example model that has been presented in table 5.7.-3.

@Risk offers you the possibility to change the graph settings according to your own wishes. You can change the scaling, labels, type of graph, etc.

All simulation data is easily transferred to a new Excel workbook for further processing. Select the Results tab in the @Risk menu bar and select the 'Results to worksheet' option. The user is then able to export all possible @Risk output.

To expand the possibilities of the exposure model you can perform several additional analyses:

1. Sensitivity analysis: This kind of analysis gives you an overview of the relative influence of your input variables in determining the result of your output variable. An example of a sensitivity analysis is given in Figure 5.7.-4., which is based on the example described in 5.7.5.4:

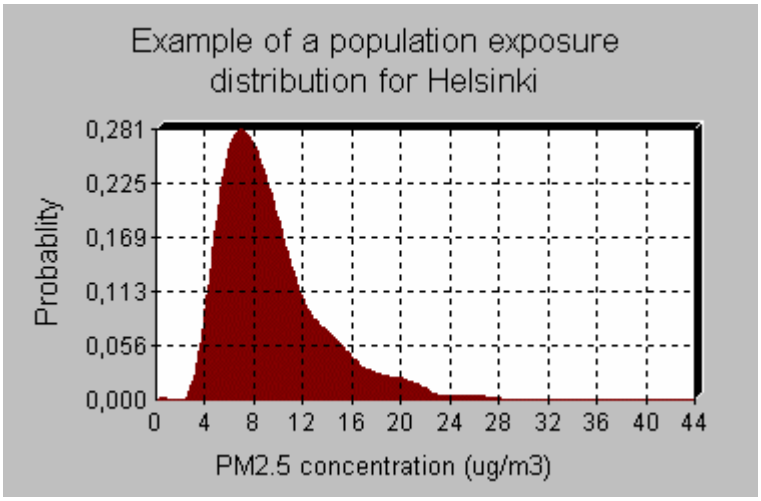
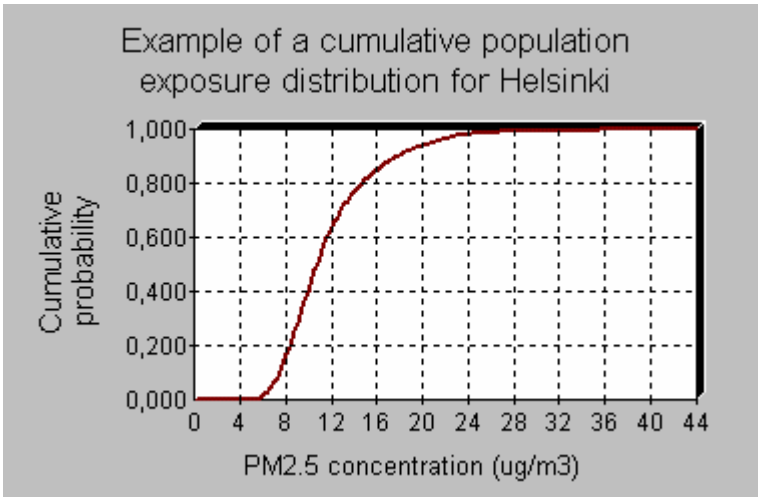


Figure 5.7.-3. Examples of @Risk output graphs (see text).



The interpretation of this graph needs some explanation. The graph shows the input distributions that affect your outcome distribution (population exposure distribution for PM2.5) most. Here we see that the inclusion of the home indoor concentration in the home µE is the most important input distribution, having a positive correlation coefficient of 0,654.. The second most influential input distribution is time spent in the home indoor microenvironment, then the time spent in the workplace and so on. 'C' points to a concentration distribution, 'f' to a distribution on time spent in a certain microenvironment, and 'S' refers to a source.

2. Scenario analysis: This analysis allows you to investigate which input distributions contribute significantly towards reaching a goal. For example, you want to know if smoking is the most influential parameter for the part of the population that has exposures to $PM_{2.5}$ higher than a predefined level.

3. Definition of target values: This gives the probability of achieving a specific outcome. For example, the percentage of the population that is exposed to concentrations of $PM_{2.5}$ higher than 50 µg/m^3.

For further information about these analysis the user is referred to the @Risk manual.

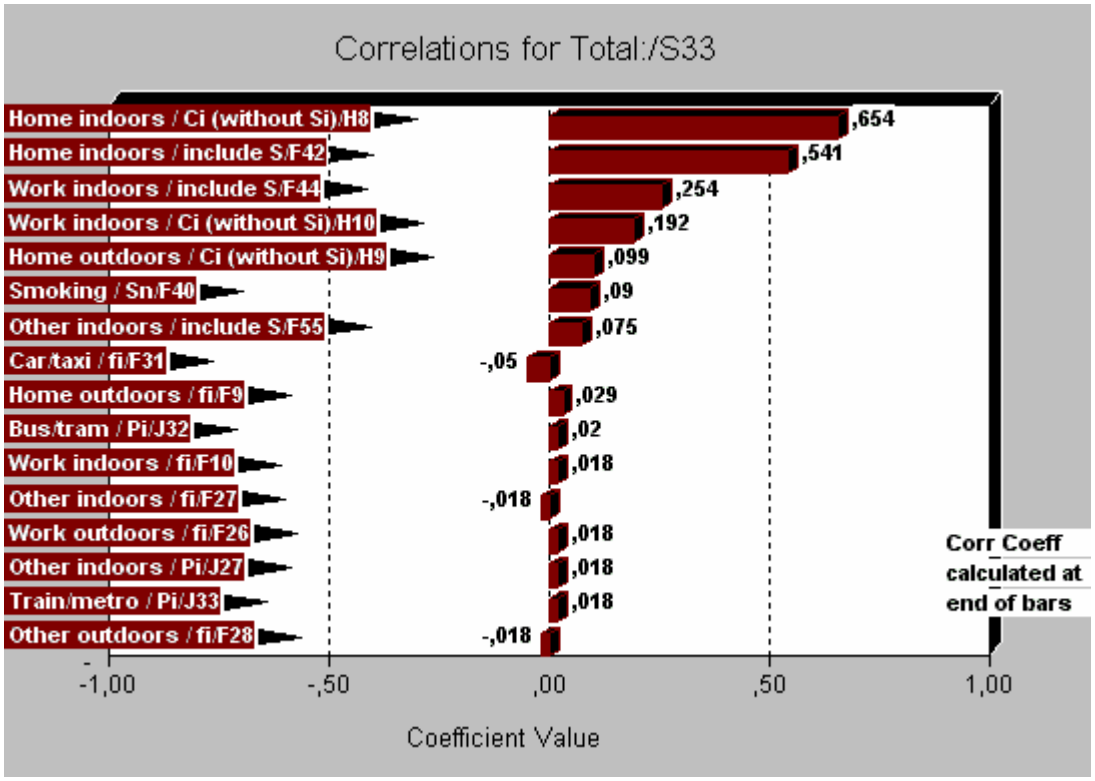


Figure 5.7.-4. Example of @Risk output for sensitivity analysis (see text)

6. DISCUSSION AND CONCLUSIONS

In general the data-analysis of the *EXPOLIS* project has just started. The initial project time was only two years, and this allowed for the technical planning, equipment testing, building up the quality assurance systems and training of the field personnel (about 6 months), field sampling, which needed to cover all seasons, i.e. a full year, and finally completion of the data entry and cleaning, which consumed the remaining time. Most of the results will be analysed and published during the two-four years to come, and consequently the following *Discussion and Conclusions* should not be viewed as presenting the final, let alone the complete results of this study. Presently this work is planned and funded in some of the *EXPOLIS* centres until year 2001, and two new *EXPOLIS* centres have been established by local funds in the U.K., namely Oxford, studied by Imperial College, and London, studied by Middlesex University.

Some Difficulties and Limitations

Some expected and unexpected difficulties in managing the field work have reduced the amount of useful data or limited its applicability from some of the *EXPOLIS* centres. We describe them here in short for the benefit of future research groups planning similar work in the future.

- The burden of the exposure monitoring and questionnaire filling tasks was considerable for the study subjects. Also, the cultural attitudes towards completing different questionnaires applied by the government, universities or market research organisations vary significantly between the different regions of Europe: In the north the public is trusting, positive and cooperative towards research for the environment and, what is perceived as common good, in the south and east the attitudes are more doubting, individualistic and much less cooperative. Consequently, obtaining a representative and cooperative population sample for a tedious field study can be a quite difficult, expensive and even impossible task. The resulting poor representativity of some of the EXPOLIS population samples might have been avoided by a multistage stratified sampling procedure combined with considerable work time allocation for each subject, i.e. more time and money. However, while poor sample representativity does affect the comparability of the measured exposure levels and distributions between the centres, it has much smaller effect on the assessments of exposure determinants and sources, or the usefulness of the data for exposure modelling purposes.
- While the field staff training and quality assurance procedures of *EXPOLIS* were extensive, they did not help in the cases, where some of the work procedures were outsourced to laboratories outside of the trained *EXPOLIS* teams, and the outside laboratories did not carefully study or follow the established work procedures and were not included in the daily technical e-mail based communication network. As a consequence some arbitrary and poorly documented changes were made in some procedures in some centres, which resulted in increased uncertainties and even some bad data that had to be identified and omitted from the results. This could have been avoided by ensuring that each laboratory and individual actually participating in the work should have been introduced to and approved by the coordinator, and be included in the training and communication network, i.e. by “increasingly bureaucratic and non-flexible project management (sic!)”.
- Local occupational safety requirements forbade some details of some operating procedures to the extent that critically reduced some data quality, and even led to data rejection. This could have been avoided by moving those procedures into another laboratory, by redesigning and testing the procedure for local use, or even cancelling that part of the work in that particular centre.

Questionnaire Data about Home, Work, Behaviour, Activity and Environment

In order to link each individual's measured exposures to indoor and outdoor environments and activites in home and workplace, commuting, other activites and personal behavior, extensive questionnaire data were collected from each studied individual using almost identical questionnaires in each centre. These data will not be discussed independently from the measured exposure data here, but because they may have also other applicability than this, the data tables presenting the basic statistics of these questionnaire results are presented in Annex II, Tables 4 - 10. We will pick only one part of the questionnaire data to closer view here, namely **perceived air pollution annoyance**, because this data is uncommon and may provide background for some interesting analyses.

Annex II, Table 11 presents the **degrees of air pollution annoyances** perceived and reported by the study subjects during the exposure monitoring period. Interestingly the degree of perceived annoyance **at home** was mostly quite low in all cities except Prague. The obvious reason for the considerably high annoyance level in Prague is the fact that the population sample there consists disproportionately of young and educated downtown dwellers. The degrees of annoyance **at work** were generally higher than at home; Grenoble, Basel and Helsinki remain low, with Athens, Prague and Milan showing a degree higher of annoyance. In general the workday ($PM_{2.5}$) exposures were indeed quite high compared to the private time exposures in Athens, but not so in Milan or Prague. In Basel the workday exposures were interestingly lower than private time exposures - they were higher in all other centres. **In commuting,** comparing the average numbers between the cities, the degree of self reported air pollution caused annoyance appears to be related to the means of commuting: Increasing share of commuting by car and taxi decreases, and increasing share of commuting by bus, tram, train or metro (i.e. public transportation) increases the degree of annoyance. This effect is less likely to be due to higher air pollution exposure in the bus or tram, but instead to higher perceived control of one's own environment in the car.

Annex II, Table 12 presents the **causes of air pollution annoyances** reported by those individuals that perceived higher than average degree of annoyance at home. **At home** "dust" was the leading cause in Helsinki, Grenoble, Prague and Milan, "exhaust gases" were perceived most important in Basel and "other" (environmental tobacco smoke - ETS - is the leading "other" cause) in Athens. "Chemicals" as a group was considered as the leading cause only by a few percent in any city. "Chemicals" gained some importance in **workplace** annoyance, especially in Milan, "dust" remained the leading cause in Helsinki, "exhaust gases" were perceived to be the leading cause in Prague and Basel and "other" (ETS) was considered to be most important in Athens and Grenoble.

These results need to be interpreted against the cultural and social, as well as environmental background in each city. The high proportion of "exhaust gases" annoyance in Basel is probably due to high publicity and low acceptability of this pollution source - the actual exposure to exhaust gases is probably higher in Athens, Milan and Prague. In Prague the high degree of annoyance from "exhaust gases" in the workplace can be understood against the proximity of the study population to a much debated highway tunnel project. The high proportion of annoyance caused by dust in Helsinki is not easy to explain. The $PM_{2.5}$ levels in all microenvironments and personal exposure Helsinki are clearly the lowest among the cities. "Dust" may serve as a proxy for an unidentifiable source of annoyance like the tight sealing and low ventilation rate of the buildings and very dry indoor air in the winter. The high proportion of "other" sources of annoyance in Athens and Grenoble results from ETS, in Athens because smoking is so very common, in Grenoble, because the study population consisted of asthmatics.

The causes of annoyances **in commuting** are not surprisingly dominated by "exhaust gases". Yet "dust" is considered also important in Prague and Helsinki - probably due to different reasons - and "other" in Athens, again probably due to cigarette smoke.

6.1. Exposure Frequency Distributions

$PM_{2.5}$

The $PM_{2.5}$ levels and frequency distributions in the home indoor and outdoor, and workplace air as well as personal private time and workday exposure are presented in Annex II, Table 17. The most striking features shown in this database are the very high workday exposure and workplace levels in Athens, and also the relatively high workday exposure in Prague and workplace concentration in Milan. In contrast the workday exposure in Basel is generally lower than the private time exposure. The private time exposures and also the home indoor levels are rather similar in Grenoble, Basel, Prague and Athens, but nearly 2/3 lower in Helsinki.

VOCs

Looking at some of the associations between the different target VOC compounds in the personal exposures in Helsinki (Annex II, Table 18/H);

- trimethylbenzene is best correlated with TVOC,
- nonane, decane and undecane are highly correlated with each other, indicating a common source, which is not outdoor air, where the levels are much lower than at exposure,
- ethylbenzene, o,m,p-xylenes and trimethylbenzene are closely correlated with each other, but not with the previous group, also indicating a common source, again not outdoors air where the levels are much lower than at exposure,
- benzene - for which a new European air quality directive is established - is poorly correlated with both of the previous groups, somewhat better correlated with toluene. Also the benzene levels are higher in indoor than outdoor air, and
- TVOC levels (judged by the geometric mean, median, and 90^{th} percentiles) are the highest in personal exposure and home indoor air, workplace levels are already lower, and home outdoor concentrations much lower. Because the TVOC concentrations vary much more than PM2.5 or CO concentrations, comparison of the arithmetic means is almost meaningless, it is so much dominated by a few very high indoor concentrations.

Comparison of the TVOCs between the cities (Annex II, Table 18/A,H,M,P) can only be done for Athens, Helsinki, Milan (no personal VOC-exposures available) and Prague. VOCs were not measured in Grenoble, and the different sampling and analytical procedures in Basel does not allow TVOC comparison, but does allow comparison of the individual compounds.

The TVOC exposures were highest in Athens, followed by Milan (estimated from microenvironmental measurements) and Prague. The mean and median TVOC exposures in Helsinki were about 1/3 to 2/3 of the other cities. In the outdoor levels the relative differences were even higher.

The benzene levels are of particular interest, because of the new EU directive on ambient air benzene concentrations. Because the directive relates to long term benzene concentration, the most appropriate levels for comparison here are the arithmetic averages and median values, which are presented in Table 6.1.-1. for five *EXPOLIS* cities. Two things can be readily observed from the table, (i) the ambient air concentrations in Athens and Milan have clearly difficulties in meeting the benzene directive while in Basel and Helsinki even the 90^{th} percentile of the ambient air concentrations is only about 2.5 $\mu g/m^3$, and (ii) the average personal benzene exposure level is over two times higher than the outdoor air

benzene concentration, i.e. most of the benzene exposure does occur indoors and comes from indoor or personal sources.

Table 6.1.-1 Average and median benzene exposure and outdoor air concentrations in five EXPOLIS cities ($\mu g/m^3$) (values exceeding 5 $\mu g/m^3$ are marked in **bold**).

City	personal exposure level		outdoor air concentration		exposure
average	median	average	median	outdoor	
Athens	**18**	**13**	**11**	**7.8**	1.64
Basel	**5.6**	3.1	1.5	1.4	3.73
Helsinki	3.4	2.6	1.6	1.5	2.33
Milan(*	**16**	**12**	**10**	**7.4**	1.60
Prague	**12**	**9.6**	**5.2**	4.4	2.31

*) estimated from microenvironmental concentrations

6.2. Time Spent in Microenvironments

The summary statistics of the time-microenvironment-activity data from the 6 *EXPOLIS* cities (Annex II, Table 16) presents one quite surprising result, namely the fact that the total time (presented in decimal hours) spent in the traffic is so similar in all cities, in average 2 hours per day in all means of transport combined (1.88 - 2.20 h/d in the order Grenoble, Helsinki, Basel, Milan, Athens, Prague). The time spent in "private" transportation (bike, walk, motorbike, scooter, car, taxi) seems to vary even less, from 1.6 h/d in Milan to 1.8 h/d in Athens. The variations within each city are much larger than the almost nonexisting differences between the cities. The more detailed time allocations in the different means of transport vary more in relation to the size of the city, culture and availability of, e.g. metro. As an example the time spent in a car/taxi ranges from 0.46 h/d in Basel to 0.56 h/d in Prague, 0.84 h/d in Helsinki, 0.89 h/d in Milan, 1.05 h/d in Grenoble and 1.17 h/d in Athens. The average time allocation for transportation seems to be affected only little by the size of the city, the distances within it, even by the fluency of the traffic flow. One may speculate that the 2 h/d average is a result of population level optimisation between time, privacy, space and money. Given available personal funds, a desire to reduce the time spent in traffic in a large city requires normally an investment in a smaller downtown apartment with less privacy. On the other hand a desire for more private space requires an investment in a suburban home at a longer distance from work and urban services.

The average time spent at home (inside and outside) varies also little between the cities, from the lowest, 13.6 h/d, in Milan (13.9 in Basel and Helsinki, 14.3 in Prague, 14.6 in Grenoble) to the highest, 15.8 h/d in Athens. As one might expect the average time spent at work (inside and outside) progresses

almost in reverse order, from 4.4 in Athens to 5.3 in Grenoble, 5.9 in Basel and Prague, 6.1 in Helsinki and 6.6 in Milan.

The total time spent outdoors varies around 1 hour per day (0.67 - 1.20 h/d in the order of Milan, Grenoble, Prague, Helsinki, Basel, Athens), making justice to the often made claim that people spend 95 - 97 % of their time indoors. There is no obvious climate or cultural explanation to these outdoor time allocation differences; in Milan the people seem to spend very little time outdoors around the home. No other outstanding feature can be seen in this data.

Looking at the average length of reported passive exposure to tobacco smoke, the differences across Europe are distinct. The average exposure is 0.37 h/d in Helsinki, 0.58 and 0.64 h/d in Prague and Basel, 1.15 h/d in Grenoble and 1.33 and 1.36 h/d in Milan and Athens. The calculated increase of the average population exposure to $PM_{2.5}$ from the 1 hour average excess ETS exposure per day in Athens and Milan compared to Helsinki, is about 5 $\mu g/m^3$. Indeed, compared to the ambient air levels the average personal exposure levels and indoor microenvironmental concentrations are about 4 $\mu g/m^3$ higher in Athens and Milan than in Helsinki.

6.3. Exposure Determinants (Risk Factors)

The personal nighttime (daytime) exposure to $PM_{2.5}$ was clearly affected by the reported traffic density near home (workplace) and the general type of home (workplace) outdoor environment in Athens and Basel and Helsinki. Because of the long exposure times at home and in work, this result was expected. An unexpected result is that time spent in different means of private (car, motorcycle) and public (bus, tram, metro, train) urban transport showed no consistent impact on $PM_{2.5}$ exposures in any of the *EXPOLIS* cities. Several different reasons could together explain this unexpected result: The time spent in traffic is in the average only 2 hours per day, and its direct impact on the 24 h day exposure is therefore limited. Increasing time (and exposure) in traffic is usually associated with a home location in a suburban or rural location, and thus somewhat lower nighttime exposure. And finally the urban air $PM_{2.5}$ pollution from traffic (and other sources) is smoothly distributed over large areas, i.e. although much of our urban exposure comes from traffic sources, most of it is indirect, occurring at home, in the workplace and also outdoors in the form of traffic generated particles that have dispersed over the entire city. When the personal $PM_{2.5}$ exposures were compared directly against the ambient air concentrations of black smoke (BS) in Athens and PM10 in Basel and Helsinki, the association was in general poor. However, when the same comparison is done for nighttime exposure only against ambient air PM2.5 in Helsinki, the association improves, and when the ETS exposed subjects are removed from the sample, the association becomes significant and correlation between personal nighttime $PM_{2.5}$ exposure and simultaneous ambient air $PM_{2.5}$ concentration increases to $r^2 = 0.70$.

Concerning the indoor sources, smoking, when occurring in the home or workplace, significantly increases one's exposure even when one does not smoke him/herself. The average increase in three cities (Athens, Basel, Helsinki) due to presence of smoking v.s. no smoking is about 20 $\mu g/m^3$, which is almost two times the average nighttime $PM_{2.5}$ exposure in Helsinki. When looking at the effect of the time of daily exposure to tobacco smoke, the increase in 24 h average exposure seems to be approximately 5 $\mu g/m^3$ for each hour of exposure to ETS. Use gas appliances in the home does not seem to have any effect on the $PM_{2.5}$ exposures, neither does the use of mechanical ventilation or air conditioning systems.

In contrast, the time that the windows are kept open in the home significantly increases the nighttime exposure levels in Helsinki and Basel. In Athens no such effect is observed. A closer analysis of this effect in Helsinki shows that the time that the windows are kept open increases indoor, and also outdoor $PM_{2.5}$ concentrations, i.e. the weather conditions when windows are kept open for long times (sunny, calm, dry, summer) are associated with higher outdoor air $PM_{2.5}$ concentrations than the weather conditions when the windows are kept closed (cloudy, windy, rainy, winter).

The determinants of other exposures than $PM_{2.5}$ are not yet analysed.

6.4. Sources of Exposure

Analysis of the sources of exposure in the *EXPOLIS* database is still very much in its infancy. Comparing the distributions of home indoor and workplace to outdoor concentrations gives indications of whether the main sources are indoors (I/O >> 1.0), both (I/O _1.0) or outdoors (I/O _1.0). Again comparing personal daytime and nighttime exposure levels with workplace and home indoor levels indicates whether the exposure sources are in those microenvironments (P/I or P/W _1.0), or elsewhere, like in commuting, other microenvironments or activities (P/I or P/W > 1.0). However, because the source apportionment of the samples taken and data collected in the *EXPOLIS* study, based on much more advanced data analysis methods, is already ongoing, we hesitate to assess the sources of exposure to any greater extent in this report.

6.5. European Database

The European *EXPOLIS* database containing all the measured and questionnaire collected data has been completed and will be available in 2000 from the *EXPOLIS* coordinator at the KTL Department of Environmental Hygiene in Kuopio, Finland. The database is stored in MS Access 6.0 (Office 95) format. A detailed description about the structure and contents of the database will be made openly available either in a publication or in the EXPOLIS home page or both. The actual database will be provided for any credible research group presenting a sound research proposal for the purpose of the proposal, but it will not be given for open use and wider distribution, because due to its complexity essentially infinite numbers of meaningless statistical significance tests could be made providing many statistically significant but random and meaningless correlations and test results. The delivery conditions will be set and the decision will be made by the *EXPOLIS* principal investigators.

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