Personal exposure to airborne particles

Validity of outdoor concentrations as a measure of exposure in time series studies

Nicole Janssen

Stellingen

1. In studies naar de acute effecten van 'fijn stof' in de lucht op de gezondheid kan de persoonlijke blootstelling goed gekarakteriseerd worden door middel van metingen in de buitenlucht. *Dit proefschrift.*

2. Een belangrijk deel van de zogenaamde 'personal cloud', gedefinieerd als 'een onverklaarbare verhoging in persoonlijke PM10 blootstelling ten opzichte van binnen- en buitenluchtconcentraties' (Wallace, 1996), wordt veroorzaakt door resuspensie van stof als gevolg van lichamelijke activiteit. *Dit proefschrift.*

3. De in dwarsdoorsnede studies gevonden lage correlaties tussen persoonlijke blootstelling aan stofvormige luchtverontreiniging en de concentratie in de buitenlucht (Spengler en Soczek,1984; Ozkaynak et al, 1996) zijn niet relevant voor tijdreeksstudies. *Dit proefschrift.*

4. Gezien de verschillen in de samenstelling tussen PM10 in de buitenlucht en PM10 in klaslokalen, kunnen hoge PM10 concentraties in klaslokalen niet beoordeeld worden aan de hand van advieswaarden of normen die voor PM10 in de buitenlucht zijn opgesteld. *Dit proefschrift.*

5. In onderzoek naar de *acute* gezondheidseffecten van stofvormige verontreiniging van de buitenlucht, zijn stofbronnen in de binnenlucht niet van belang.

6. De stelling dat persoonlijke stofmetingen een betere blootstellingsmaat opleveren dan stofmetingen in de buitenlucht (Mage, 1985), is niet altijd juist.

7. Het uitblijven van de realisatie van de voorgenomen klassenverkleining in het basisonderwijs heeft in het afgelopen schooljaar veel stof doen opwaaien.

8. Een uitvoering van koormuziek is leuker voor de koorleden zelf dan voor het publiek.

9. Vegetarisme wordt veelal meer gezien als een levensfase dan als een levenswijze.

10. If you're going to be able to look back on something and laugh about it, you might just as well laugh about it now. *Marie Osmond.*

11. De bal is niet altijd rond.

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Abstract

This thesis describes a study of the relation between outdoor concentrations and personal exposure to particulate matter (PM) air pollution. The main objective of the study was to examine the validity of outdoor concentrations as a measure of exposure to PM in times series studies. Repeated measurements of personal and outdoor concentrations of particles smaller than 10 μm (PM10) were conducted in 37 non-smoking adults and 45 children. In addition, repeated measurements of fine particles (FP; particles <3 μm) were conducted in 13 children. For each subject separately, personal exposures were related to outdoor concentrations using linear regression analysis. The distributions of the individual correlation coefficients were investigated. Furthermore, the extent to which differences between personal and outdoor concentrations could be explained was studied.

Personal PM10 concentrations of both adults and children were reasonably well correlated over time with ambient PM10 concentrations. Personal FP exposures were highly correlated with ambient FP concentrations. Excluding days with exposure to environmental tobacco smoke (ETS) improved the correlations. In all cases, the medians of the individual correlation coefficients were higher than the estimated cross-sectional correlations.

Personal exposures exceeded outdoor concentrations. An important part of these differences could be attributed to exposure to ETS. For non-ETS exposed subjects, differences between personal and outdoor concentrations were relatively small for PM10 in adults and for FP in children. Personal PM10 concentrations among non-ETS exposed children, however, were still more than two times higher than ambient PM10 concentrations. An important part of this remaining difference could be attributed to high PM10 concentrations in the classrooms. Results of the analysis of the elemental composition of part of the classroom PM10 samples suggest that these high classroom concentrations were due to resuspension of coarse particles and/or suspension of soil material.

The findings of this study provide support for the use of fixed site measurements as a measure of exposure to PM in epidemiological time series studies linking the day-to-day variation in PM to the day-to-day variation in health endpoints.

Contents

1. Introduction

Background

In air pollution epidemiology, exposure variables used in practice usually are surrogates or proxies of the 'true' exposure of the study subjects. The validity and precision with which the 'true' exposure is being approximated may vary widely from one exposure variable to the next. Estimates of the relationship between exposure and health effect can be severely biased when exposure is assessed inaccurately and/or imprecisely[1-3]. To evaluate the validity of a specific exposure variable, information about the correlation between this variable and the 'true' exposure is necessary. In practice, however, a perfect exposure measure is generally not available. In this case, the exposure measure used must be compared with an exposure measure, which is considered a more accurate approximation of the 'true' exposure[4].

Exposure to a pollutant is defined as the event when a person comes into contact with a pollutant of a certain concentration during a certain period of time[5]. This definition distinguishes exposure from concentration on the one hand and dose on the other hand. A concentration is a quantitative expression of the presence of a pollutant, but there is no exposure unless there is physical contact with human beings. A dose, on the other hand, refers to the amount of pollution that actually crosses one of the body's boundaries[3]. Air pollution levels can show substantial spatial and temporal variation. When people move from one location to the other during the day, they can therefore encounter different concentrations during various time periods. This is taken into account by the concept of integrated personal exposure (E):

$$E = \int_{t1}^{t2} C(t) \times dt$$

where C(t) is the air pollution concentration, which varies over time period t1 to t2 with increment dt[1].

The integrated personal exposure can be assessed directly or indirectly. In the direct approach, measurements are conducted using personal monitors that are attached near the subjects' breathing zone. In the indirect approach, measurements at different microenvironments are combined with data on time

activity. The general form of the equation used to calculate this time-weighted integrated exposure is:

$$E_i = \sum_{j}^{J} C_j \times t_{ij}$$

where C_j is the concentration in microenvironment j during the time period t_{ij} that individual i remained in microenvironment j and J is the total number of microenvironments[1,6]. A microenvironment is defined as a three-dimensional space where the pollutant level at some specified time is uniform or has constant statistical properties[6]. In practice, the indirect approach involves measurements in a few selected microenvironments that are considered to have a major contribution to the integrated exposure[1]. Direct measurements of personal exposure are often considered the most accurate estimate of the subject's true exposure[2,6], but are also the most expensive and intrusive.

In the last decade, a large number of epidemiological studies have been published documenting effects of several air pollutants on health[7]. Recently, especially concern about the effects of particulate matter (PM) air pollution has increased. Epidemiological studies have documented associations between PM air pollution and several acute health effects, including mortality, hospital admissions, respiratory symptoms and lung function[7-9]. These studies are mostly time series studies, relating day-to-day variations in air pollution to day-to-day variations in health endpoints. In these studies, exposure assessment is generally based on measurements conducted on fixed sites in ambient air. Measurements of personal exposure are considered a more accurate estimate of the subject's true exposure[10]. To investigate the validity of ambient concentrations measured at a fixed site as a measure of exposure, therefore, information about the correlation between these fixed site measurements and measurements of personal exposure, as well as information about potential differences in concentration levels, is necessary. It has been suggested that PM concentrations measured at fixed sites in ambient air correlate poorly with personal exposures[11], which raises questions about the plausibility of the observed exposure-response relationships[12]. In most personal exposure studies, however, the correlation between personal and outdoor concentrations was calculated cross-sectionally: personal exposure data were collected from a group of subjects by measuring different subsets of subjects on different days (=different ambient concentrations) and measuring each subject once or only a

limited number of times. Next, one correlation coefficient was calculated, using all measurements from all subjects and days. This correlation is influenced by the variation in personal exposure between subjects. Since time series studies relate day-to-day variations in outdoor concentrations to day-to-day variations of health endpoints, the correlation between personal and ambient concentrations within persons, over time, is more relevant than the variation between persons. This correlation may be better because factors that can cause variation between subjects, such as exposure to environmental tobacco smoke (ETS), are less variable in time within subjects, and therefore mainly cause variation between subjects. At present, only limited information is available about the within-subject correlation between personal and outdoor PM concentrations[13,14].

Characteristics of particulate matter air pollution

Particulate matter air pollution refers to an air-suspended mixture of solid and liquid particles that vary in size, composition, and origin[8]. Particle size is often expressed in terms of the aerodynamic equivalent diameter, defined as the diameter of a unit-density sphere having the same gravitational settling velocity in air as the particle in question[15]. The size distribution of particles in ambient air consists of modes (peaks in the distribution), which can be described by lognormal functions[16]. Basically, there are two different modes: fine mode particles, which are formed by condensation of gases or vapours, and coarse mode particles, which are generated through mechanical processes[8].

The fine mode can consist of at least two sub-modes: the nucleation mode and the accumulation mode. The nucleation mode, also termed 'ultrafine particles', consists of particles with diameters less then ~0.08 μm that are emitted directly from combustion processes or that condense from gases after emission[17]. Coagulation increases the particle size, but nucleation mode particles do not tend to grow over into the size range of the accumulation mode. Instead, nucleation mode particles move into the accumulation mode by coagulation with accumulation mode particles, this being favoured over coagulation with other nucleation mode particles because of the greater surface area of the larger particles. Nucleation mode particles have a relatively short lifetime and are detected only in the vicinity of particle emitting sources or when new particles have been recently formed in the atmosphere[16,17]. The accumulation mode consists of particles with diameters between 0.08 and ~2 μm[17]. These particles

are formed by gas-to-particle conversion through chemical reactions and condensation as well as coagulation. The rate of particle growth through these processes slows down with increasing particle diameter. As a result, the accumulation mode does not extend much beyond a few micrometers in diameter, and remains distinct from the larger particles in the coarse mode[16].

The coarse mode consists primarily of particles generated by mechanical processes. This mode contains mainly windblown dust, sea salt spray, and plant material[16]. Basically, coarse mode particles result from a size reduction of larger particles[15]. However, as particles become smaller, more and more energy is required to break them into smaller units. This establishes a lower limit of approximately 1 μm for coarse particles[18]. Generally, particles less than 100 μm are considered to stay airborne long enough to be observed and measured as aerosols[15], thus defining the upper limit of the coarse particle mode. The dividing line between fine and coarse is usually taken to be at about 2 μm particle diameter, which is the minimum in the mass distribution between the accumulation and coarse mode[16]. Fine and coarse particles may overlap in the intermodal region between 1 and 3 μm[18].

Due to the different sources and processes of formation, fine and coarse mode particles do not only differ in size but also in composition. Fine mode particles consist primarily of sulphates, nitrates, ammonium and organic and elemental carbon, whereas coarse mode particles are largely composed of oxides of crustal material (such as silicon, aluminium, titanium and iron), sea salt and plant material[16-18]. In addition, fine and coarse mode particles also differ in, among others, atmospheric half-life, deposition rates and thus travel distance: fine mode particles can travel 100s to 1000s of kilometres, whereas coarse mode particles can travel for less than 1 to 10 kilometres[18]. Furthermore, particle size influences the deposition in the respiratory system: particles smaller than 2-3 μm can penetrate into the gas-exchange region of the respiratory tract[19], whereas larger particles are more likely to deposit in the upper airways or larger lower airways[8].

Exposure measurements of ambient particulate matter air pollution

Particulate matter air pollution concentrations are usually defined as the amount of mass in a unit volume of air ($\mu g/m^3$)[6,15] . Much of the early work relating health effects to particulate measurements, however, was done using data from the

1950s and 1960s in the UK, when the measurements were carried out using the 'black smoke' method[1]. This method involves the collection of particles on a paper filter after which the reflectance of the sample is measured and transformed into $\mu g/cm^2$ using an equation describing the Standard Smoke curve[20]. Since this transformation is based on empirical curves determined in the early 1960s, when airborne particle concentrations were dominated by soot from incomplete coal combustion, black smoke concentrations nowadays are considered unreliable as a measure of mass concentrations and should not be compared directly with gravimetrically measured concentrations[1,21,22].

Epidemiological studies often use measurements available from regulatory monitoring networks, set up to determine compliance with air quality standards, as a measure of exposure. Many epidemiological studies of air pollution in the 1960s and 1970s in the USA therefore used TSP (total suspended particulates) measurements, measured by high-volume samplers, as the indicator of particle exposure[8]. The particle sizes collected with this sampling method were poorly defined and the upper 50% cut-point was found to range from 25-50 μm, dependent on wind speed and direction[17,22]. In the late 1970s and early 1980s, the US Environmental Protection Agency (EPA) established a network of measurements of fine and coarse particles using dichotomous samplers with cut points of 15 μm and 2.5 μm[8,18]. The 15 μm cut point was chosen to measure 'inhalable' particles, to define the fraction of particles which can primarily deposit in the conducting airways and gas-exchange areas of the human respiratory system during mouth breathing. The second cut point of 2.5 μm was based upon considerations of the chemical composition and size distribution of the particles, and on the predominant penetration of particles $\leq$ 2.5 μm into the gas-exchange region of the respiratory tract[19]. The 15 μm cut point subsequently was changed to 10 μm to measure the thoracic fraction, i.e. the particles that penetrate through the larynx and are available for deposition on the tracheobronchial and/or the alveolar epithelia[22]. Detailed information about particulate sampling methods and related issues can be found in a review by Chow[17].

In 1987, a National Ambient Air Quality Standard for PM10 (i.e. particulate matter with a 50% cutoff diameter of 10 μm) was promulgated in the United States, to replace the earlier TSP standard. The WHO Air Quality Guidelines for Europe[23] of 1987 also include a PM10 guideline. Recent epidemiological studies have therefore often used PM10 measurements as the basis of exposure estimation[8]. More recently, however, it has been suggested that fine particles are more likely to be responsible for the observed associations between PM air

pollution and respiratory health effects[18,24]. For example, Schwartz *et al.*[25] found that fine particles, measured as PM2.5, and not coarse particles (PM10 minus PM2.5) are specifically associated with mortality. In the United States, therefore, in the new air quality standards for particulate matter, the existing standards for PM10 were recently supplemented with PM2.5 limit values[26]. In the European community, although no PM2.5 standard will be established, PM2.5 monitoring will be mandated. It can therefore be expected that future studies will include PM2.5 measurements for exposure estimation.

Personal exposure to particulate matter air pollution

A recent review of studies on personal and indoor particle concentrations is given by Wallace[13]. Personal exposure studies conducted in the 1970s-1980s generally measured respirable suspended particles (RSP), defined as particles with a 50% cut off of 3.5 μm, which at the time was the American Conference of Governmental Industrial Hygienists (ACGIH) conventionalised alveolar fraction[27,28]. Most of these studies showed poor (cross-sectional) correlations of personal exposures with outdoor concentrations[9]. For example, Sexton *et al.*[29] and Spengler *et al.*[30] found values of 0.06 and 0.07, respectively, for the correlation between personal and ambient RSP. At the start of the study presented in this thesis, only two studies on personal exposure to PM10 had been published: The Particle Total Exposure Assessment Methodology (PTEAM) study[31,32], conducted in Riverside, California, in 1990, and the Total Human Environmental Exposure Study (THEES)[14,33], conducted in Phillipsburg, New Jersey, in 1988.

In the PTEAM study, personal measurements of PM10 were conducted for two consecutive 12-hour periods on 178 non-smoking subjects. Each subject was measured once and up to 4 subjects were measured each day. Concurrently, indoor and outdoor measurements of both PM10 and PM2.5 were conducted at each home. In addition, outdoor measurements were conducted at a fixed monitoring site. Personal exposures were only moderately correlated with outdoor concentrations: the (cross-sectional) correlation between personal PM10 concentrations and fixed site outdoor concentrations was 0.37 for daytime samples, 0.54 for nighttime samples and 0.42 for 24-hour averaged concentrations. The cross-sectional correlations between personal and indoor concentrations were higher: 0.63, 0.88 and 0.74 for daytime, nighttime and 24-hour averaged concentrations respectively[31,32,34,13].

In the THEES study, 24-hour averaged personal sampling of PM10 was conducted on 14 non-smoking adults for 14 consecutive days. Indoor sampling was conducted in the homes of the subjects (8 homes) and outdoor monitoring was conducted at 4 sites throughout the study area. Buckley *et al.*[14] calculated the correlation between personal and outdoor PM10 for each subject individually, using 9 to 14 personal measurements from 13 of the subjects. The median of the individual correlation coefficients was 0.53 (range 0.14 to 0.90). Correlating the personal concentrations with a time weighted average of indoor and outdoor concentrations did not improve the correlations (median R = 0.55). Lioy *et al.*[33] reported the indoor – outdoor relationships of the eight homes in the THEES study. The cross-sectional correlation was 0.67 (n = 101), whereas individual correlation coefficients ranged from 0.64 to 0.98. This study suggests that correlations between personal or indoor and outdoor concentrations, within subjects or homes, are indeed higher than cross-sectional correlation coefficients.

Another observation from most personal exposure studies is that personal PM concentrations are generally higher than indoor or outdoor concentrations[13]. Cigarette smoking is considered to be the most important source of excess personal or indoor particle concentrations. Wallace[13] estimated that the increase of PM2.5 concentrations in homes with smokers ranges from 25 to 45 $\mu g/m^3$; the contribution of a single cigarette was estimated to range from 1 to 2 $\mu g/m^3$, averaged over a 24-hour period[13].

In the PTEAM study, the average personal PM10 concentration during daytime was 150 $\mu g/m^3$; about 60% higher than the average indoor and outdoor concentrations of 95 $\mu g/m^3$. During nighttime, the average personal concentration was much lower (77 $\mu g/m^3$) and more comparable to the average indoor (63 $\mu g/m^3$) and outdoor concentrations (mean 86 $\mu g/m^3$)[32]. Although exposure to ETS was found to significantly increase indoor concentrations and night-time personal concentrations, the average daytime personal concentrations did not significantly differ between subjects exposed to ETS (mean 155 $\mu g/m^3$) and non-ETS exposed subjects (mean 147 $\mu g/m^3$)[32], suggesting that the excess personal exposures were caused by other factors. Resuspension of particles by personal activities and proximity to particles-generating sources have been suggested as causes of the so-called 'personal cloud'[13].

If the sources of excess personal exposure are constant (within a person in time) this will only result in a systematic difference between personal and outdoor concentrations but will not influence the correlation between the concentrations

over time. In this case the power of a study to detect a relationship between exposure and disease is not compromised. In quantitative terms, however, the detected relationship can still be biased[3]. If the sources of excess exposure are less constant (within a person in time), the correlation between personal and outdoor concentrations will generally be reduced. This is demonstrated in the THEES study: data on daily activity were collected and these activity variables were included (via stepwise regression) in a model that related personal concentrations with time weighted averages of indoor and outdoor concentrations. The correlation improved for all subjects to a median R of 0.93 (range 0.58 to 0.999). House-cleaning activities, cooking, use of unvented kerosene space heaters and ETS exposure were found to be especially important[14]. In addition to the influence of particle sources on the correlation between personal and outdoor concentrations, there may be differences in the composition of the particles that are biologically relevant[3]. The sources of excess personal exposures therefore need to be better understood.

Goals of the study:

The goals of the study were:

1. To evaluate the relation between personal and ambient airborne particulate matter (PM) concentrations, within subjects, over time
2. To evaluate potential differences between personal, indoor and ambient PM concentrations

Study Design

A personal exposure study was conducted in which repeated measurements of personal and outdoor PM were conducted, to allow calculation of the correlation within subjects, over time. The personal exposure study was conducted within the framework of a panel study on acute effects of air pollution on respiratory health in the Netherlands[35]. Averaging time, particle size, and population selection were linked to the design of this panel study. This implied 24-hour averaged measurements of PM10 in groups of 50- to 70-year-old and 10- to 12-year-old primary school children, living in the city of Amsterdam and in the small town of Wageningen. For adults, only non-smoking subjects with no smokers in

their households were included in the study. Adults were measured in the winter and fall of 1994. Children were measured in the winter and spring of 1994 and 1995. Considering the recent attention for fine particles, measurements of FP were added to the study in the spring of 1995 (children in Wageningen only). Information about factors that might influence exposures was obtained by questionnaire. In the homes of the adults, repeated measurements of indoor PM10 were added to provide information about the relation between personal and indoor concentrations, and the relation between indoor and outdoor concentrations as well. In addition, indoor measurements were conducted in the classrooms of the children.

Structure of the thesis

In chapter 2, the methods used to measure the personal exposure to PM10 and FP are described. Chapters 3, 4 and 5 describe the relation between personal and outdoor concentrations of PM10 in a group of adults (chapter 3), PM10 in a group of children (chapter 4) and FP in a group of children (chapter 5), respectively. Correlations within subjects, over time, as well as an evaluation of the differences in concentration levels are described. One of the findings of the study on childhood exposure to PM10 (chapter 4) was that PM10 concentrations in classrooms are considerably higher than outdoor concentrations, causing large differences between personal and outdoor PM10 concentrations in children. In chapter 6, the causes of these high classroom PM10 concentrations are further investigated. Finally, chapter 7 presents a general discussion of the most important findings and implications.

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2. Personal sampling of airborne particles: method performance and data quality*

Nicole A.H. Janssen, Gerard Hoek, Hendrik Harssema and Bert Brunekreef

Abstract

A study of personal exposure to inhalable particles (PM10) and fine particles (FP) was conducted in groups of 50- to 70-year-old adults and primary school children in the Netherlands. Four to eight personal measurements per subject were conducted, on weekdays only. Averaging time was 24 hours. Method performance was evaluated regarding compliance, flow, weighing procedure, field blanks and co-located operation of the personal samplers with stationary methods. Furthermore, the possibility that subjects change their behaviour due to the wearing of personal sampling equipment was studied by comparing time activity on days of personal sampling with time activity on other weekdays. Compliance was high; 95% of the subjects who agreed to continue participating after the first measurement successfully completed the study and, except for the first two days of FP sampling, over 90% of all personal measurements were successful. All pre and post sampling flow readings were within 10% of the required flow rate of 4 l/min. For PM10, precision of the gravimetric analyses was 2.8 $\mu g/m^3$ and 0.7 $\mu g/m^3$ for filters weighted on an analytical and a micro-balance respectively. The detection limit was 10.8 $\mu g/m^3$ and 8.6 $\mu g/m^3$ respectively. For FP, weighing precision was 0.4 $\mu g/m^3$ and the detection limit was 5.3 $\mu g/m^3$. All measurements were above the detection limit. Co-located operation of the personal sampler with stationary samplers gave highly correlated concentrations ($R>0.90$). Outdoor PM10 concentrations measured with the personal sampler were on average 4% higher compared to a Sierra Anderson (SA) inlet and 9% higher compared to a PM10 Harvard Impactor (HI). With the FP cyclone 6% higher classroom concentrations were measured compared to a PM2.5 HI. Adults spent significantly less time outdoors (0.5 hour) and more time at home (0.9 hour) on days of personal sampling compared to other weekdays. For children no significant differences in time activity were found.

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Introduction

In air pollution epidemiology, exposure assessment is traditionally based on fixed site measurements in ambient air. However, measurements of the personal exposure are considered a more accurate estimate of the subject's true exposure[1]. In order to investigate the validity of ambient concentrations measured at a fixed site as a measure of exposure to air pollutants, therefore, information about the correlation between fixed site measurements and measurements of personal exposure is necessary.

For particles, instruments that are available for personal air sampling have generally been developed for use in occupational settings and are not necessarily suited for use in the general environment. Limitations exist, among others, in terms of noise, battery life-time, detection limit and the possibility of interference with normal daily activities. Studies on personal exposures to particles in the general environment, therefore, have needed to find ways to overcome these limitations. To reduce pump noise levels, for example, Thomas *et al.*[2] added noise damping material in the pump and Lioy *et al.*[3] packed the pump in an acoustic shell. Solutions for insufficient battery life-time include changing the batteries after 12 hours[3] or plugging the pumps into the nearest wall socket when possible[4].

In the winter and fall of 1994 and 1995 a personal exposure study was conducted among 10- to 12-year-old children and 50- to 70-year-old adults in the Netherlands. The main objective of this study was to conduct repeated measurements of personal and outdoor particles, to allow calculation of the correlation between personal and outdoor concentrations within subjects over time. Repeated measurements of PM10, and to a smaller extent fine particles (FP), were conducted. This paper describes the methodologies and performance of the personal PM10 and FP sampling and analysis methods. Results of the analyses of the relationship between personal and outdoor concentrations will be published elsewhere.

Methods

Study design

The personal exposure study was conducted within the framework of a panel study on acute effects of air pollution on respiratory health[5]. Population selection, particle size and averaging time were linked up with the design of the panel study, implying 24-hour averaged measurements of PM10 in groups of 50- to 70-year-old adults and primary school children. Considering the recent attention for fine particles, measurements of FP were added in the second year of the study (children only). Seven to eight personal measurements per subject were planned, on weekdays only. Measurements were spaced approximately one week apart. 50- to 70-year-old adults, living in Amsterdam, and 10- to 12-year-old children, living in Amsterdam and Wageningen, were invited to participate in the study. For adults, measurements took place from 17 January to 31 March 1994 and from 17 October to 23 December 1994. Children were measured from 16 February to 19 April 1994 (1 school) and from 11 January to 15 June 1995 (4 schools).

Outdoor concentrations were obtained from fixed monitoring sites. For adults, outdoor measurements were conducted using an inlet similar to the Sierra Anderson 241 dichotomous sampler inlet[6]. For children, both in Amsterdam and in Wageningen, outdoor measurements were conducted using the same samplers as used for personal sampling.

Indoor measurements of PM10 were conducted in the living rooms of the adults (both periods) and in the children's classrooms (1995 only), using a Harvard Impactor (HI)[7,8]. In the classroom of the children who participated in the study on fine particles, indoor PM2.5 measurements were conducted, using a PM2.5 HI.

Sampling equipment

Personal measurements of PM10 were conducted using a personal impactor described by Buckley *et al.*[9] (A.D.E Inc., Naples, Maine, USA). 25 mm 3 μm pore size diameter Gelman Teflon filters (Gelman R2PI025) were used. Air was sampled at 4 l/min using a flow-controlled battery operated pump (Gillian, model Gil-Air 5). Pump noise levels were reduced by placing the pump in an acoustic shell, consisting of a silencer - placed on both the inlet and the outlet of the pump - and a plastic cover lined with insulation material (figure 1). Adults could

wear the monitor in a made-to-fit bag with a belt and shoulder strap. This bag was transformed into a backpack when worn by children. The bag with the monitor weighed approximately 1.5 kg. The impactor was attached near the breathing zone, to (the collar of) the subject's clothing or the shoulder strap of the bag. At night the bag with the pump was placed near the bed in a wooden insulated box to further reduce pump noise levels (figure 2). Furthermore, the box contained a wall plug convertor, which subjects had to connect to the pump before they went to sleep, to reduce battery usage over night, allowing the pump to run for 24 hours on one charge. The impactor was attached to the outside of the box (figure 3).

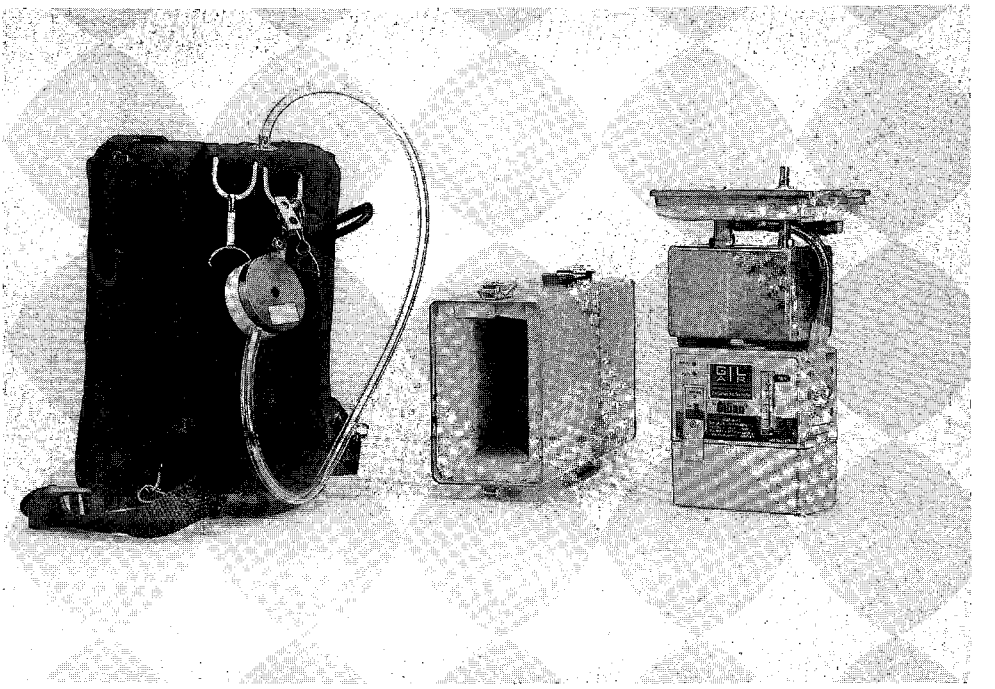

Figure 1. Different parts of the personal monitor: right: pump with silencer; middle: insulation cover; left: complete monitor

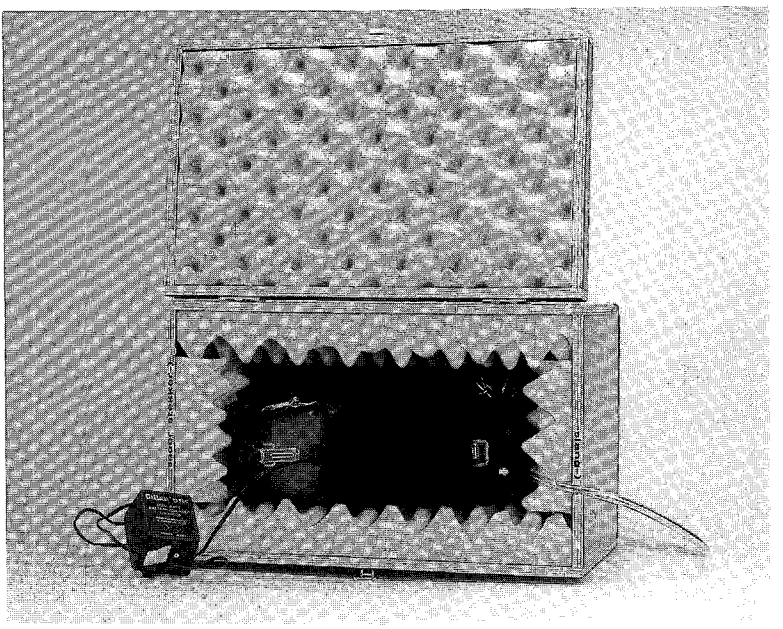

Figure 2. View from above of the monitor in the 'night-box' (left: wall plug convertor)

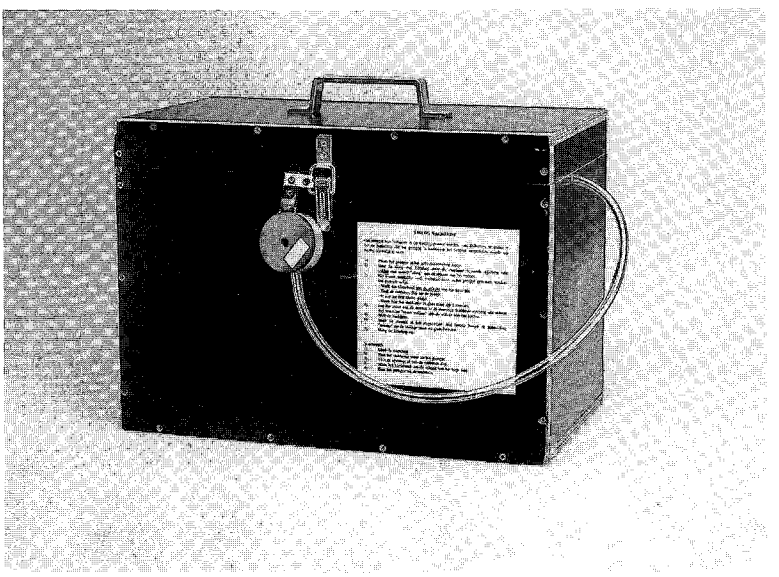

Figure 3. Front view of the 'night-box', with the impactor attached to the outside of the box

Measurements of fine particles were conducted using the Casella respirable dust cyclone (Casella Ltd. London, UK). This cyclone is usually used at a flow rate of 1.9 l/min to measure particles with a 50% cut-off (D_{50}) of 5 μm in occupational studies. For the aim of this study, however, a smaller cut-off was desired, which can be realized by sampling at a higher flow rate. According to Ogden *et al.*[10] the flow dependence of the Casella cyclone can be described as:
$D_{50} = 8 \times Q^{-0.715}$ where Q = flow rate (l/min)
This implies that a flow rate of 5 l/min would be necessary to sample PM2.5, which could not be achieved with the pumps available. Because in the personal PM10 study the pumps had proved to perform well for 24 hours at 4 l/min, it was decided to use the same flow rate for the FP sampling. At this flow rate, the cyclone is expected to measure particles with a 50% cut-off of about 3 μm. Except for the sampling head, the same equipment and filters were used as for PM10 sampling.

Sample collection
For the adults, samplers were distributed and collected at the homes of the participants. During the first home visit participants received specific instructions on how to wear the sampler. For the children the samplers were distributed and collected at school, except for the first measurements when samplers were distributed at the children's homes to effect individual instruction of the children in the presence of one of the parents.

Participants were instructed to wear the sampler whenever possible, but they were allowed to place the sampler nearby - with the impactor attached to the bag, oriented in the same way as when it was worn (figure 1, left) - during activities in which wearing the sampler would be too inconvenient (e.g. sports) or impossible (swimming). Adults and parents were asked to record the kind and duration of those activities as well as the position of the sampler during these activities.

Flows were measured at the beginning and end of each 24-h sampling period with calibrated rotameters and elapsed time indicators were used to calculate the sampled volumes. In some cases, the pump had stopped running before the end of the 24-hour sampling period. Consistent with specifications of the manufacturer, a laboratory experiment, which involved measuring the flow of 5 pumps repeatedly until they stopped because of battery failure, did not show a stronger decrease in flow near the end of the battery life-time. In case

no post flow was available, therefore, the post flow was estimated by subtracting the average flow difference (pre minus post) of all full-time measurements from the pre-flow. Measurements that had lasted less than 20 hours were excluded.

Gravimetric analysis

Prior to weighing, filters were stored in a refrigerator at 4 °C. Filters were weighed after equilibrating at about 20° C and 44% RH for 24 hours, using desiccators. All filters collected in 1994 were weighed using an analytical balance with 10 μg reading precision; filters collected in 1995 were weighed on a micro-balance (Mettler, type MT5) with 1 μg reading precision. All filters were weighed twice, on different days and the average of the two filters weights was used in calculations. When the difference between two weights of PM10 filters equalled or exceeded 50 μg, the filter was weighed a third time. For the weighing of FP filters, which were all conducted on the micro-balance, filters were reweighed when duplicate weighings were more than 10 μg apart. In the case of triplicate weighing, the weight was calculated as the average of those two weights within the allowed range of 50 or 10 μg. When the third weight was within this range for both the first and the second weighing the average of all three weights was used.

Field blanks of personal measurements were prepared by assembling filters in the impactor or cyclone and carrying these samplers to the participants' homes and schools along with the samplers used for measurements. All samplers were transported in sealed plastic containers. Mean field blank weight changes were subtracted from all sample weights.

Field comparison

The personal PM10 impactor was co-located with a Sierra Anderson sampler and a PM10 HI on the outdoor monitoring site in Amsterdam, on 24 and 15 days respectively, including all days of personal sampling of children in Amsterdam. The cyclone used for the FP measurements was co-located with a PM2.5 HI in a classroom on 12 days.

The relationship between the measurement methods was assessed using a method suggested by Cornbleet and Gochman[11]. This method simultaneously minimizes the squared distances from the observed data points to the regression line in the horizontal and vertical direction. One regression line is obtained

regardless of which of the two methods is considered as the independent variable. This method was used instead of ordinary least squares regression because it is not obvious which variable should be selected as the independent variable. In addition bias of the 'true' regression slope to the null occurs in ordinary least squares regression when a considerable amount of measurements error is present[11]. Slope and intercept were calculated using the formulas given by Cornbleet and Gochman[11]. We assumed that the absolute error of the two compared methods was the same.

Interference with normal time activity

After each day of measurements, adults and parents were asked to fill out a questionnaire including questions about the time they had spent in several micro-environments during the 24 hour sampling period. Furthermore, during the 2-3 months measuring period, participants kept a daily diary which among others contained questions on time spent outdoors (all subjects) and time spent at home (from October 1994 onward). This information was used to investigate the possibility that subjects changed their behaviour due to the wearing of personal sampling equipment, by tending to stay at home or spending less time outdoors. For each subject the average time spent outdoors and time spent at home was calculated for sampling days and non-sampling days separately. Since personal measurements were conducted on weekdays only, weekends were excluded in the calculation of the non-sampling days' time activity. Furthermore, holidays and days subjects reported to have spent at home because of illness were excluded. For each subject the difference between time spent outdoors or time spent at home on sampling days and non-sampling days was calculated. The distribution of these individual differences was investigated. The hypothesis that the mean difference is zero was tested using the Wilcoxon signed rank test.

Results

Compliance

51 adults agreed to participate. After the first measurement, subjects were explicitly asked whether they were sure they were able to wear the monitor another 7 days, after which 12 adults decided to drop out. Of the remaining 39 adults, 37 subjects successfully completed the study; the other 2 adults were

excluded because of non-compliance. 63 children were included in the study. 1 child dropped out after the first measurement, 3 children were excluded because of non-compliance and 1 child was excluded because she had changed home during the period of measurements. The study was successfully completed by the remaining 58 children, of whom 45 were involved in the PM10 study and 13 in the FP study.

For the adults and children who successfully completed the study, over 90% of the personal PM10 measurements succeeded; 262 of out 290 (90.3%) for adults and 301 out of 333 (90.4%) for children. 60 measurements (9.6%) were lost, due to pump failure (30 times, 4.8%), negligence of using the wall plug convertor overnight (11 times, 1.8%), battery failure within 20 hours (9 times, 1.4%), filter damage (5 times, 0.8%) and other causes (5 times, 0.8%). Pump failure implied that the pump had stopped operating due to another cause than battery failure. Pump failure occurred automatically when the pump could not maintain it's flow, for example due to blocking of the sampling tube for one minute.

For the FP measurements, a high percentage (72%) of the filters of the first two days of measurements were damaged, caused by small irregularities in the filter holder. Replacing the filter screen and an adjustment in the assembling procedure solved this problem. On the remaining 6 days of sampling, 92.1% of the conducted measurements succeeded, resulting in a total number of 77 successful personal FP measurements.

Flow

All pre and post sampling flow readings were within 10% of the required flow rate of 4 l/min. The average flow rate was 4.00 l/min (sd: 0.08; range 3.82 to 4.26). Flows slightly decreased during the 24 hour measuring period; the average difference between pre and post flow (pre minus post) was 0.11 l/min (sd 0.09; range -0.12 to 0.42). This mean difference was used to extrapolate the post flow in case the pump had stopped running before the end of the 24 hour measuring period. This extrapolation was necessary in 9.9% of the adults' measurements and 6.9% of the children's measurements. The frequency of empty batteries increased during the course of the study. For example, in the first group of children it happened in 3.3% of the measurements compared to 10.4% in the fourth (last) group of children.

Gravimetric analysis

Triplicate weighing of personal PM10 filters was necessary in 9.7% of all weighings conducted on the analytical balance. For the PM10 filters that were weighed on the micro-balance, all duplicates were within the range of 50 μg so no triplicate weighing was conducted. Precision of one weight determination, calculated as the mean coefficient of variation of duplicates within the 50 μg criterium times the mean filter weight, was 11.6 μg for the analytical balance and 3.0 μg for the micro balance. Since the calculation of the sampled mass involves a subtraction of two weight determinations, this implies a precision of the mass determination of 16.4 ($\sqrt{(11.6^2 + 11.6^2)}$) for the analytical balance and 4.2 μg for the micro balance. With the sampled volume of 5.8 m^3 that is 2.8 $\mu g/m^3$ and 0.7 $\mu g/m^3$ respectively.

Triplicate weighing of personal FP filters was conducted in 16.9% of all weighings. Precision of the mass determination for duplicates within the 10 μg criterium was 2.5 μg, i.e. 0.4 $\mu g/m^3$.

Field blank values and detection limit

The mean mass increase on field blanks of personal PM10 samples was 26.1 μg (n = 27; sd 20.8; range −5 to 100 μg) for filters weighed with the analytical balance and 30.8 μg (n = 11; sd 16.6; range 7 to 55.5 μg) for field blanks weighed with the micro-balance. The detection limit, defined as 3 times the standard deviation in field blanks divided by the sampled volume of 5.8 m^3, was 10.8 $\mu g/m^3$ for the analytical balance and 8.6 $\mu g/m^3$ or the micro-balance. The mean mass increase on field blanks of personal FP samples was 9.3 μg (n = 7; sd 10.1; range 0.5 to 31 μg), resulting in a detection limit of 5.2 $\mu g/m^3$. All PM10 and FP measurements were above the detection limits.

Field comparison

Results of the comparison of the personal PM10 sampler (PS) with the Sierra Andersen sampler (SA) and the PM10 Harvard Impactor (HI) are presented graphically in figure 4. The estimated regression equations were: PS = 4.6 + 0.89 x SA (R = 0.95; n = 24) and PS = 0.1 + 1.09 x HI (R = 0.91; n = 15). The mean difference in concentrations was +0.8 $\mu g/m^3$ (sd 6.0) for the difference between the personal and the outdoor impactor (PS minus SA) and +2.8 $\mu g/m^3$ (sd 4.4) for the difference between the personal and indoor sampler (PS minus

HI); the mean percentage difference was +3.9% (sd 16.1) and +9.5% (sd 16.5) respectively.

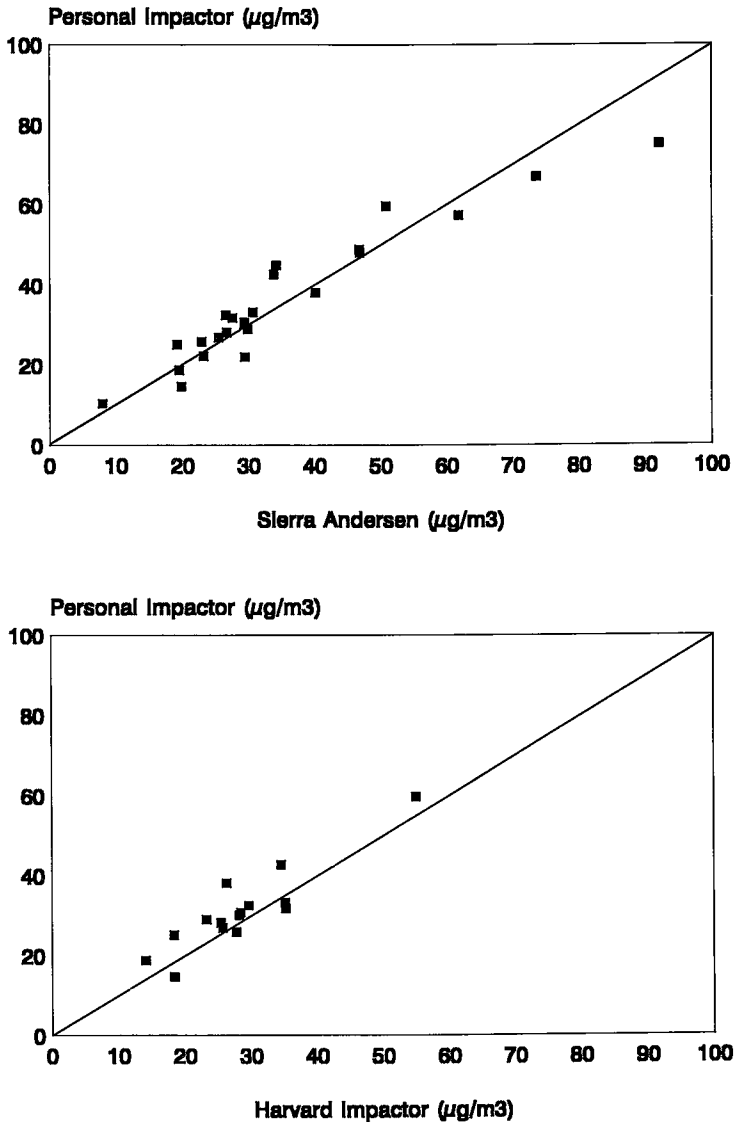

Figure 4. PM10 concentrations in outdoor air measured with a personal PM10 impactor compared to a SA sampler (above) and a PM10 HI (below) (line shows 1:1 line)

Results of the comparison of the Casella cyclone (CC) and a PM2.5 HI are given in figure 5. The estimated regression equation was: CC = 0.6 + 1.01 x HI (R=0.96; n=12). The mean difference between the cyclone and the impactor (CC minus HI) was +0.8 $\mu g/m^3$ (sd 1.7); the mean percentage difference was +6.4% (sd 11.8).

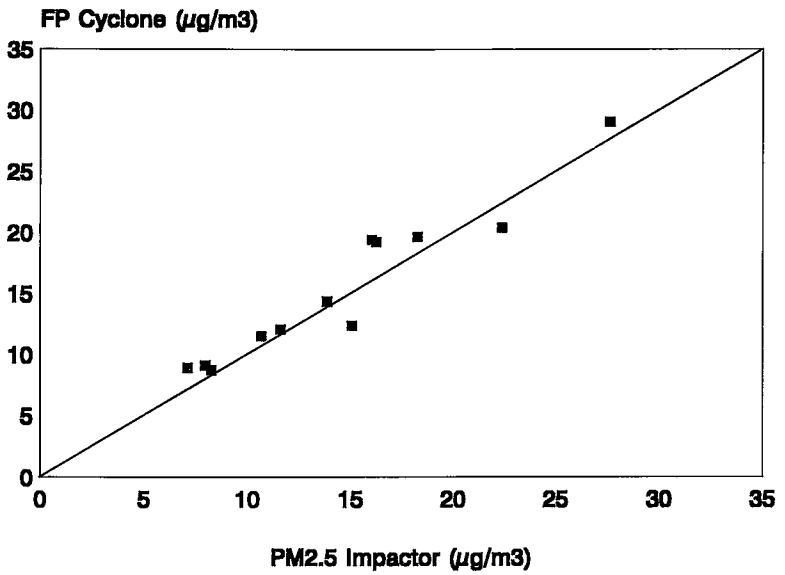

Figure 5. Classroom FP concentrations measured with a personal cyclone compared to PM2.5 concentrations measured with a PM2.5 HI (line shows 1:1 line)

Interference with normal time activity

The distributions of the individual averages of time spent outdoors and time spent at home on days of personal sampling and on non-sampling days are presented in table 1. Three children were excluded because of incomplete or incorrect completion of the daily diary. The number of observations per subject ranged from four to eight for sampling days and 25 to 54 for non-sampling days. Adults spent significantly less time outdoors and more time at home on days of personal sampling compared to other weekdays. For children, no significant differences were found.

Table 1. Distribution of individual averages of time (hours) spent outdoors and at home on days of personal sampling and non-sampling days

	Adults				Children			
	n	Mean	sd	Range	n	Mean	sd	Range
Time spent outdoors								
Sampling days	37	1.3	0.8	0.1 to 3.4	55	2.8	0.9	1.0 to 5.0
Other days	37	1.8	0.9	0.5 to 4.0	55	2.9	0.8	1.1 to 4.6
Difference	37	-0.5**	0.6	-2.3 to 0.4	55	-0.1	0.6	-1.2 to 1.5
Time spent at home								
Sampling days	24	20.5	2.7	11.3 to 23.8	40	14.7	1.0	11.7 to 17.4
Other days	24	19.6	2.4	11.9 to 22.8	40	14.9	0.9	12.4 to 16.9
Difference	24	0.9*	1.6	-1.6 to 3.9	40	-0.2	1.0	-1.6 to 2.0

* *Wilcoxon signed rank test mean = 0; $p < 0.05$*

** *Wilcoxon signed rank test mean = 0; $p < 0.01$*

n *number of subjects*

Apart from sleeping, taking showers and getting dressed, the average time that subjects recorded not to have carried the pump was 0.4 hours per measurement for adults and 1.0 hours per measurement for children. During the major part of these activities (76% for adults and 84% for children) the monitor was placed in the subject's vicinity. Only occasionally subjects recorded to have left the pump at home when they went outside their own home (19 times for adults and 31 times for children; on average 2.0 hours per occasion), mostly during sports or social and cultural activities such as parties, funerals, cinema or theatre visits. However, the number of times subjects took the pump with them and placed it nearby during similar activities was higher; 32 times for adults and 148 times for children (on average 1.4 hours per occasion).

Discussion

The main objective of this study was to calculate the correlation between personal and ambient particles within subjects over time. For this purpose, repeated measurements were necessary. Because conducting personal measurements is very labour intensive, it was important to minimize drop-out during the course of the study. After the first measurement, therefore, participants were explicitly asked whether they were willing to carry the monitor another 7 days. This resulted in the drop-out of a considerable number of adults (24%), but resulted in a high compliance among the remaining volunteers: 95% of the subjects who agreed to continue participating after the first measurement, both adults and children, successfully completed the study.

Except for the first two days of FP sampling, less than 10% of all conducted personal measurements were lost, half of which were caused by pump failure. Only 11 samples (1.8%) were lost because subjects had forgotten to use (or incorrectly used) the wall plug convertor overnight. This shows the feasibility of asking subjects to conduct these kind of operations themselves, instead of sending out field technicians to change the battery at half-time, as was done in a study by Lioy *et al.*[3] However, 9 measurements (1.4%) were lost due to battery failure within 20 hours, and during another 52 measurements the pump had stopped running before the end of the full 24-hour measuring period. This implies that, for some subjects, solely the use of a wall plug convertor overnight is not sufficient to prolong the battery life-time to 24 hours, possibly because the time they sleep is not long enough to sufficiently re-charge or relieve the battery. The problem increased during the course of the study, suggesting that aging of the pumps plays a role. Sexton *et al.*[4] realized a 24 hour sampling period on one battery by plugging the monitor into the nearest wall socket whenever possible (e.g. during indoor sedentary activities). In our study we wanted subjects to carry the sampler as much as possible, because due to potential variations within the same room, for example caused by sources such as smokers, the concentration in the breathing zone can differ from the concentration at the site were the monitor would be placed. In future studies the problem of how to realize a desired sampling time longer than can be achieved on one battery charge, needs further attention.

The procedures for the gravimetric analysis included duplicate weighings of all filters and conducting a third weighing in case of an unacceptable variation

in the duplicate. This procedure was especially important in the first phase of the study, when filters were weighed on an analytical balance with 10 μg reading. Furthermore, outliers caused by errors in the weighing procedure were avoided. For PM10, the precision of the mass determination was 2.8 $\mu g/m^3$ for the analytical balance and 0.7 $\mu g/m^3$ for the micro-balance. For adults, who had lower personal exposure than children and whose filters were all weighed on the analytical balance, the average personal exposure was 62 $\mu g/m^3$, so the error introduced into the concentration by the weighing procedure was 4.5%. For FP all weighings were conducted on a micro balance. The precision was 0.4 $\mu g/m^3$; i.e. 1.4%, when divided by the average personal FP concentration of 28 $\mu g/m^3$.

Field blanks showed a mean mass increase of 26.1 μg (sd 20.8) for filters weighed using an analytical balance; field blanks weighed on a micro-balance even showed a somewhat higher mean mass increase of 30.8 μg (sd 16.6), ruling out the gravimetric analysis as an explanation for the blank values. The mean mass increase of field blanks of FP samples was considerably lower, 9.3 μg (sd 10.1), suggesting the problem is related specifically to the PM10 impactor. All components of the impactor were thoroughly washed before each use. The assembling procedure involves several operations, during which contamination of the filters might have occurred. However, all concentrations were above the detection limits, so the mass increase did not result in unmeasurable values.

Concentrations measured with the personal samplers were highly correlated ($R>0.90$) with concentrations measured with stationary methods at the same monitoring site. On average, the personal PM10 sampler gave higher concentrations compared to a Sierra Anderson inlet (4%) and a Harvard PM10 IASI (9%). These differences are within the range of differences found in other particle measurement comparison studies[12]. Classroom concentrations measured with the FP cyclone, which is expected to measure particles with a 50% cut-off of about 3 μm, were on average only 6% (and not significantly) higher compared to a PM2.5 HI. This is in line with the general mass distribution of particles in ambient air, where - because of the low quantities of particles in the 1 to 3 μm size range - a small shift in cut-point near 2.5 μm will only have a small effect on the mass collected[12].

Adults spent significantly less time outdoors and more time at home on days of personal sampling, compared to other weekdays. For children, no significant differences in time activity were found, possibly because of the

classwise approach: all participating children from one class wore the sampler on the same day, so carrying a monitor did not make them exceptional. The absolute differences in the adults' time activity on days of personal sampling compared to other weekdays were rather small; on average +0.9 hours for time spent at home and -0.5 hours for time spent outdoors. For particles, it is not probable that such a small shift in time activity will cause large differences in the exposure measured. However, for other air pollutants, such as ozone, the relative shift of 32% in time spent outdoors may cause significant underestimation of the actual exposure. Furthermore, only the change in time spent outdoors and time spent at home could be evaluated. It is not known to what extent other aspects of behaviour are influenced. In personal exposure studies in general, therefore, the possibility that subjects change their behaviour due to the carrying of personal sampling equipment needs to be recognized and reckoned with.

In summary, this study has shown that conducting repeated 24 hour averaged measurements of personal exposures to particles among both children and older adults is feasible. Compliance among our volunteers was high. Quality of the measurements, indicated by data completeness, flow stability, precision of the gravimetric analysis and limit of detection, was satisfactory. The difference in time spent outdoors and time spent at home between days of personal sampling and other weekdays observed in the adults, however, shows that there is a true possibility that subjects change their behaviour due to the wearing of personal sampling equipment.

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3. Personal sampling of PM10 in adults: relation between personal, indoor and outdoor concentrations*

Nicole A.H. Janssen, Gerard Hoek, Bert Brunekreef, Hendrik Harssema, Iwan Mensink and Arjan Zuidhof

Abstract

To investigate the validity of outdoor PM10 concentrations as a measure of exposure in time series studies, the association between personal and outdoor concentrations, within subjects, over time was investigated. Repeated measurements of personal, indoor and outdoor PM10 were conducted among 37 non-smoking, 50- to 70-year-old adults, living in Amsterdam, the Netherlands, 1994. Regression analyses were conducted for each subject separately and the distribution of the individual regression and correlation coefficients was investigated. Furthermore, the extent to which differences between personal, indoor and outdoor concentrations could be explained was studied. The median Pearson's R between personal and outdoor concentrations was 0.50. Excluding days with exposure to environmental tobacco smoke (ETS) improved the correlation to a median R of 0.71. The estimated cross-sectional correlations were lower; 0.34 and 0.50, respectively. Outdoor concentrations (mean 42 $\mu g/m^3$) exceeded indoor concentrations (mean 35 $\mu g/m^3$) but underestimated personal exposures (mean 62 $\mu g/m^3$). The major part of the difference between personal and outdoor concentrations could be attributed to exposure to ETS, living along a busy road and time spent in a vehicle. The results show a reasonably high correlation between personal and outdoor PM10 within individuals, providing support for the use of ambient PM10 concentrations as a measure of exposure in epidemiological studies linking the day-to-day variation in particulate matter air pollution to the day-to-day variation in health endpoints such as mortality, hospital admissions, respiratory symptoms and lung function.

* *American Journal of Epidemiology 1998;147:537-547.*

Introduction

Recent epidemiological studies have documented associations between particulate matter (PM) air pollution and several acute health effects, including mortality, hospital admissions, respiratory symptoms and lung function[1-7]. These studies are mostly time series studies, relating day-to-day variation in air pollution to day-to-day variation in health endpoints. In these studies, exposure assessment is based on fixed site measurements in ambient air. It has been suggested that PM concentrations from fixed sites correlate poorly with personal exposures[8]. Sexton *et al.*[9] and Spengler *et al.*[10] found values of 0.06 and 0.07, respectively, for the correlation between personal and ambient respirable suspended particulates (RSP). More recently, in the Particle Total Exposure Assessment Methodology (PTEAM) study, the correlation between 24-hour averaged personal and ambient PM10 was 0.48[11]. If the variation in outdoor levels of particulate matter is not tightly linked to variation in personal exposures, the use of outdoor concentrations as a surrogate for personal exposures would tend to misclassify personal exposures and exposure-response relationships could be attenuated[12]. However, in most personal exposure studies, the correlation between personal and outdoor concentrations is calculated cross-sectionally. Personal exposure data are collected from a group of subjects by measuring different subsets of subjects on different days (=different ambient concentrations) and measuring each subject a limited number of times. Next, one correlation coefficient between personal and ambient concentrations is calculated, using all measurements from all subjects and days. This correlation is influenced by the variation in personal exposure between subjects. Since time series studies relate day-to-day variations in outdoor concentrations to day-to-day variations of health endpoints, the correlation between personal and ambient concentrations within persons, over time, is more relevant than the variation between persons. This correlation may be better because some aspects that can cause variation between subjects, such as smoking habits, are less variable in time within subjects, and therefore mainly cause variation between subjects. At present, only limited information is available about the within-subject correlation between personal and outdoor PM10 concentrations[11,13].

To investigate the validity of outdoor concentrations as a measure of exposure to PM10 in time series studies, information about the correlation between personal and outdoor measurements within subjects is necessary. We

therefore conducted a personal exposure study in which repeated measurements of personal and outdoor PM10 were conducted, to allow calculation of the correlation within subjects, over time. In addition, repeated measurements of indoor PM10 were conducted to provide information about the personal-indoor and indoor-outdoor correlations as well. This paper describes the relationship between personal, indoor and outdoor PM10 concentrations in a group of 50- to 70-year-old Dutch adults.

Materials and methods

Study design

The personal exposure study was conducted within the framework of a panel study on acute effects of air pollution on respiratory health[14]. This study was partly conducted in Amsterdam, the capital of the Netherlands, which has about 720,000 inhabitants. The major sources of air quality are local traffic and long distance transport. A number of Amsterdam subjects, who had agreed to participate in the panel study, were invited to participate in the personal exposure study. Interested non-smoking subjects with no smokers in their households and no occupational exposure to dust received a detailed written description of the study and were then asked for final consent after approximately one week. Of 195 adults approached, 51 (26%) both met the selection criteria and agreed to participate. After the first measurement, subjects were explicitly asked whether they were sure they were able to wear the monitor another 7 times, after which 12 subjects decided to drop out. A total of 37 of the 39 remaining subjects successfully completed the study.

Measurements took place in two periods: from 17 January to 31 March, 1994, involving 13 adults, and from 17 October to 23 December, 1994, involving another 24 adults. 24-hour averaged measurements of personal and indoor PM10 were conducted simultaneously, on weekdays only. One to 12 subjects were monitored on the same day and for each subject measurements were spaced approximately one week apart. Samplers were distributed and collected at the homes of the participants between 9:00 a.m. and 6:00 p.m. Seven to eight personal measurements per subject were planned. In the first period, indoor measurements were scheduled on only about five days of personal sampling because of limited indoor sampling equipment availability. In the

second period, indoor measurements were conducted on all days of personal sampling. Outdoor concentrations of PM10 were obtained from a fixed monitoring site (see below).

Information on general characteristics such as housing conditions was assessed by questionnaire. In addition, participants were asked to fill out a more detailed questionnaire including questions on exposure to ETS, time spent in several micro-environments, cleaning and cooking activities, etcetera, after each individual day of personal measurements. Exposure to ETS was assessed by means of the following questions:

1) Has anybody smoked in your living room during the measurements? yes / no
   1a) If yes, how much? _____cigarettes/cigars/pipes
2) Have you been in a room, other than your own living room, where people smoked? yes / no
   2a) If yes, how long did you stay there? _____hours

Sampling methods

Personal measurements were conducted using a personal impactor described by Buckley *et al.*[13], using 25 mm diameter 3 μm pore size Gelman Teflon filters (Gelman Sciences, Ann Arbor, Michigan) and a flow-controlled battery operated pump (model Gil-Air 5; Gilian Instruments Corp., West Caldwell, New Jersey) at a flow rate of 4 l/min. Details about the sampling method and quality issues are described elsewhere[15].

Measurements of PM10 indoors were made with a Harvard impactor (HI) (A.D.E., Inc., Naples, Maine) operating at 10 l/min[16,17], using a flow controlled pump (model SP-280E, A.D.E Inc.), using Anderson 37 mm 2 μm pore size Teflon filters (Gelman Sciences). Indoor samples were taken in the living room at a height of 1.5 m.

Outdoor PM10 concentrations were obtained from a fixed monitoring site operated for the panel study mentioned earlier[14]. The site was located in a park in the city center, about 150 m away from the nearest busy road and away from local particle sources, such as construction work or industrial sources. At this site, measurements were conducted at 1.5 m height on a continuous, daily basis (from 3 p.m. to 3 p.m.), using an inlet similar to the Sierra Anderson (SA) 241 dichotomous sampler inlet[18] at a flow rate of 16.7 l/min. Co-located operation of the personal sampler (PS) with the outdoor sampler (SA) and the indoor sampler

(HI) at the outdoor monitoring site did no show significant differences in outdoor concentrations obtained with the different methods. The estimated regression equations were PS = 4.6 + 0.89 x SA. (R=0.95) and PS = 0.1 + 1.09 x HI (R=0.91)[15]. The personal impactor was oriented in the same way as when it was worn during personal sampling.

For logistical reasons it was not possible to start the personal and indoor measurements at the same time as the outdoor measurements. The average overlap between the measuring periods of personal/indoor and outdoor samples was 21 hours. For 95% of the measurements the overlap was larger than 18.9 hours.

For all three types of measurements (personal, indoor and outdoor), flows were measured at the beginning and end of each 24-hour sampling period with calibrated rotameters, and elapsed time indicators were used to calculate the sampled volumes.

Filters were weighed using a Sartorius model 1712 (Sartorius AG, Goettingen, Germany) (first period) or Mettler model AT261 (Mettler-Toledo, Greifensee, Switzerland) (second period) analytical balance with 10 μg reading, after equilibrating at about 20°C and 44 percent relative humidity for 24 hours, using desiccators. All personal filters were weighed in duplicate[15]. Mean field blank weight changes were 26.1 μg (n=27; sd 20.8) for the personal filters, 0.4 μg (n=14; sd 31.7) for the indoor filters and 58.7 μg (n=30; sd 77.5) for the outdoor filters. These mean values were subtracted from the respective sample weights. Detection limits, defined as 3 times the standard deviation of field blanks divided by the sampled volume, were 10.8 $\mu g/m^3$, 6.6 $\mu g/m^3$ and 9.7 $\mu g/m^3$ for the personal, indoor and outdoor measurements, respectively.

Data Analysis

Correlation between personal, indoor and outdoor PM10 concentrations.

The correlation between personal, indoor and outdoor PM10 concentrations was assessed by means of individual regression analysis, using the SAS (Statistical Analysis System) procedures "PROC REG" and "PROC CORR". The following models were used:

model 1: $PM10_{personal,\,i\,t} = \alpha_{i\,1} + \beta_{i\,1} \times PM10_{outdoors,\,t}$

model 2: $PM10_{personal,\,i\,t} = \alpha_{i\,2} + \beta_{i\,2} \times PM10_{indoors,\,i\,t}$

model 3: $PM10_{indoors,\,i\,t} = \alpha_{i\,3} + \beta_{i\,3} \times PM10_{outdoors,\,t}$

Where i=subject i, t=day t and 1,2,3=model 1,2,3 respectively

The distribution of the individual regression results was investigated. Medians are presented because most correlation and regression coefficients were not normally distributed (Shapiro-Wilk Statistic, $p<0.05$). Although all subjects were non-smokers, not living with smokers, participants could still be exposed to ETS elsewhere, or at home in the case of a smoking visitor. To investigate the influence of occasional exposure to ETS on the relationship between personal, indoor and outdoor PM10, the same regression analyses were conducted after excluding days with exposure to ETS. Subjects with less than four remaining observations were excluded.

For comparison purposes, we calculated what the correlation would have been, if it had been calculated cross-sectionally. In this analysis, we randomly selected one measurement per subject and next calculated the cross-sectional correlation between personal and outdoor concentrations. This procedure was repeated 1,000 times, and the median of those 1,000 correlation coefficients was calculated to get a more reliable estimate of the cross-sectional correlation.

Difference between personal, indoor and outdoor PM10 concentrations.

The questionnaire data were used to examine to what extent differences between personal, indoor and ambient concentrations could be explained by certain characteristics or activities, such as exposure to ETS. The difference between personal and outdoor concentrations or the difference between indoor and outdoor concentrations was used as the dependent variable in a regression analysis. The SAS procedure "PROC MIXED" was used to adjust regression results for correlations between repeated measurements. A random intercept model was used. In the analysis of the difference between indoor and outdoor concentrations, cooking was considered separately for homes with and without a kitchen in open connection with the living room (a so-called "open" kitchen). Different questions on cleaning activities (dusting, vacuum cleaning, sweeping and cleaning a pet's cage) were combined into one variable 'cleaning activities'.

Results

Population

A total of 37 adults, 18 males and 19 females, successfully completed the study. The average age was 62 (range 51 to 70) years. Ten subjects (27

percent) were still employed, of whom three were teachers, two had office jobs, one was a house-painter, three worked at home and one (saleswoman) worked only one day per week. On the days of personal measurements, subjects spent on average 1.3 hours outdoors and 20.5 hours at home. One married couple participated in the study; therefore, indoor measurements were conducted in 36 houses.

All subjects lived in the inner city, within a radius of 5 km and at at most 4 km distance from the outdoor monitoring site. Seven (19 percent) subjects lived along a busy road, defined as living in a street that was part of the Amsterdam main road network. The average number of cars passing through these seven streets was 13,500 per day (range 7,125 to 17,093); for trucks the average was 670 per day (range 307 to 1,086). The mean ambient temperature during the sampling period was 6 °C.

Particle concentrations

From each adult, five to eight personal concentrations and four to nine indoor concentrations were obtained. The distributions of the individual averages of personal, indoor and outdoor PM10 are presented in table 1. Outdoor concentrations exceeded indoor concentrations but considerably underestimated personal exposures. This will be discussed in more detail later.

Table 1. Distribution of individual averages of personal, indoor and outdoor PM10 concentrations from 50- to 70-year-old adults

			PM10-concentrations ($\mu g/m^3$)			
	n	(#*)	Median	Mean	(Sd)	Range
Personal	37	(262)	56.4	61.7	(18.3)	38.0 to 112.8
Outdoor	37	(285)	41.5	41.5	(4.3)	31.9 to 50.2
Indoor	37	(254)	34.7	35.1	(9.3)	18.6 to 65.3
Difference personal-outdoor	37	(262)	15.9	20.4	(17.9)	-6.4 to 68.8
Difference personal-indoor	37	(231)	22.4	26.9	(20.7)	-1.0 to 99.9
Difference indoor – outdoor	37	(254)	-10.5	- 6.7	(9.4)	-20.3 to 15.2

* *Total number of observations*

Correlation between personal, indoor and outdoor PM10 concentrations

Results from the individual regression analyses with all observations are presented in table 2 and figure 1. Median Pearson's R was 0.50 for model 1 (personal-outdoor), 0.72 for model 2 (personal-indoor) and 0.73 for model 3 (indoor-outdoor). After excluding days with exposure to ETS (table 3; figure 2), median correlation coefficients increased and median intercepts decreased. For model 1 and 2, only 23 of the 37 subjects were included in table 3 because the other 14 subjects did not have at least four days of measurements without exposure to ETS. For model 3, only days with exposure to ETS inside the subject's own home were excluded, after which 32 homes had at least four remaining observations. For model 1 and 2, 16 of the 23 subjects included in table 3 were not exposed to ETS on any of the days of measurements, so only seven subjects had different regression results in table 3 when compared with those in table 2. For those seven subjects, after excluding days with exposure to ETS, median Pearson's R increased from 0.50 to 0.81 for model 1 and from 0.69 to 0.78 for model 2. For model 3, only three homes had different regression results. All three homes had higher Pearson's R's after excluding days with exposure to ETS.

*Table 2. Distribution of individual regression results of personal, indoor and outdoor PM10 concentrations from 50- to 70-year-old adults**

	Model 1 (n=37)		Model 2 (n=37)		Model 3 (n=36)	
	$PM10_{personal} = PM10_{outdoors}$		$PM10_{personal} = PM10_{indoors}$		$PM10_{indoors} = PM10_{outdoors}$	
	Median	Range	Median	Range	Median	Range
Intercept ($\mu g/m^3$)	32.7	-22.6 to 95.0	30.4	-138.6 to 64.7	11.5	-63.6 to 55.4
Slope	0.53	-0.40 to 2.08	0.90	- 0.16 to 6.12	0.47	-0.10 to 2.62
Pearson's R	0.50	-0.41 to 0.92	0.72	- 0.10 to 0.98	0.73	-0.88 to 0.95

* *All median intercepts, regression and correlation coefficients are significant (signed rank-test; $p<0.01$)*

*Table 3. Distribution of individual regression results of personal, indoor and outdoor PM10 concentrations from 50-to 70-year-old adults, after excluding days with exposure to ETS**

	Model 1 (n=23) $PM10_{personal} = PM10_{outdoors}$		Model 2 (n=23) $PM10_{personal} = PM10_{indoors}$		Model 3 (n=32) $PM10_{indoors} = PM10_{outdoors}$	
	Median	Range	Median	Range	Median	Range
Intercept ($\mu g/m^3$)	27.2	-22.6 to 82.5	13.1	-16.3 to 62.1	11.5	-63.6 to 35.2
Slope	0.55	-0.37 to 2.08	1.00	- 0.16 to 2.24	0.47	-0.04 to 2.62
Pearson's R	0.71	-0.41 to 0.94	0.86	- 0.10 to 0.98	0.75	-0.11 to 0.92

* *All median intercepts, regression and correlation coefficients are significant (signed rank-test; $p<0.01$)*

The average range per subject in outdoor concentrations (maximum minus minimum) on days of personal measurements was 48.4 $\mu g/m^3$ (sd 11.4; Range 24 to 64 $\mu g/m^3$). Excluding the five subjects with the smallest range (i.e. < 35 $\mu g/m^3$) did not substantially change the medians or ranges of the correlation and regression coefficients. For example, the median correlation between personal and outdoor concentrations after the exclusion was 0.51, compared 0.50 for all subjects.

Janssen *et al.*[15] reported that these adults spent significantly less time outdoors and more time at home on days of personal sampling compared to other weekdays. The differences ranged from -2.3 to +0.4 hours (mean -0.5 hours) for time spent outdoors and from -1.6 to +3.9 hours (mean +0.9 hours) for time spent at home. To investigate whether this change in behavior had any influence on the relationship between personal and outdoor/indoor PM10, the mean differences were used to divide the subjects into two groups, and the distributions of the regression results per group were calculated. No considerable differences between the two groups were found. For example, the median Pearson's R between personal and outdoor concentrations was 0.47 for subjects who spent >0.5 hours less time outdoors on days of personal measurements compared to other weekdays and 0.51 for subjects with smaller differences between time spent outdoors on days of personal measurements and other weekdays.

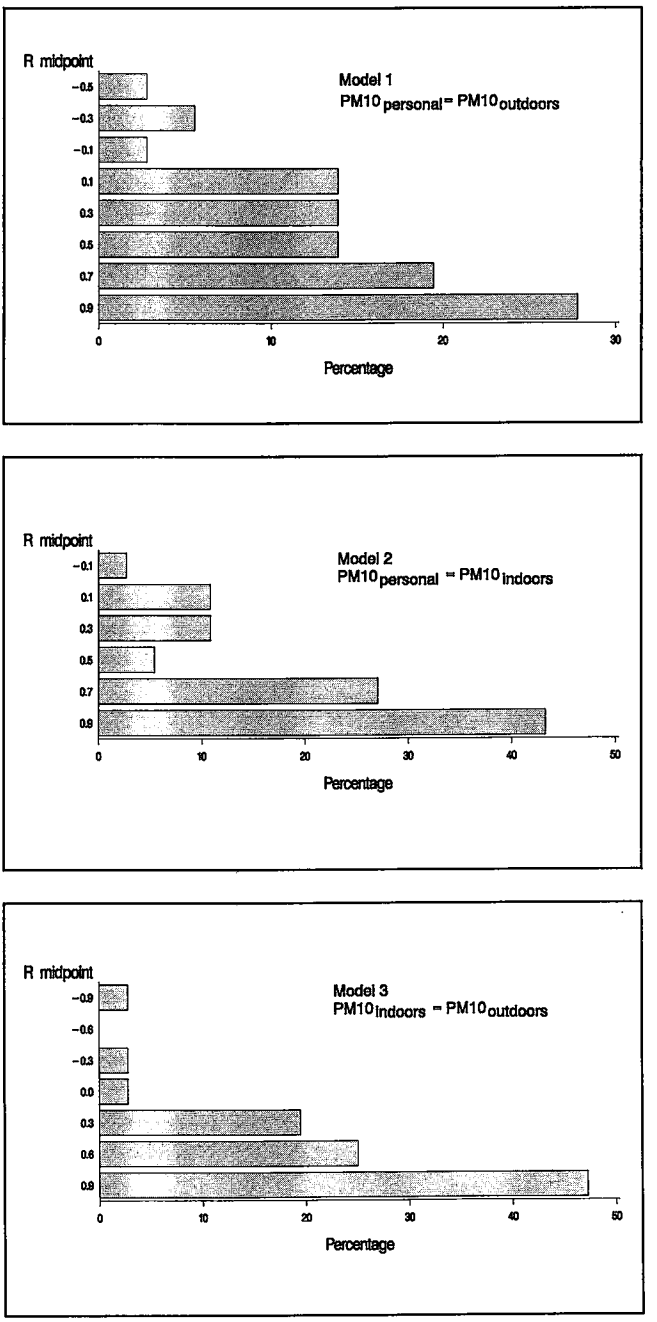

Figure 1. Distribution of individual Pearson's correlation coefficients from 50- to 70-year-old adults (n = 37 for models 1 and 2; n = 36 for model 3)

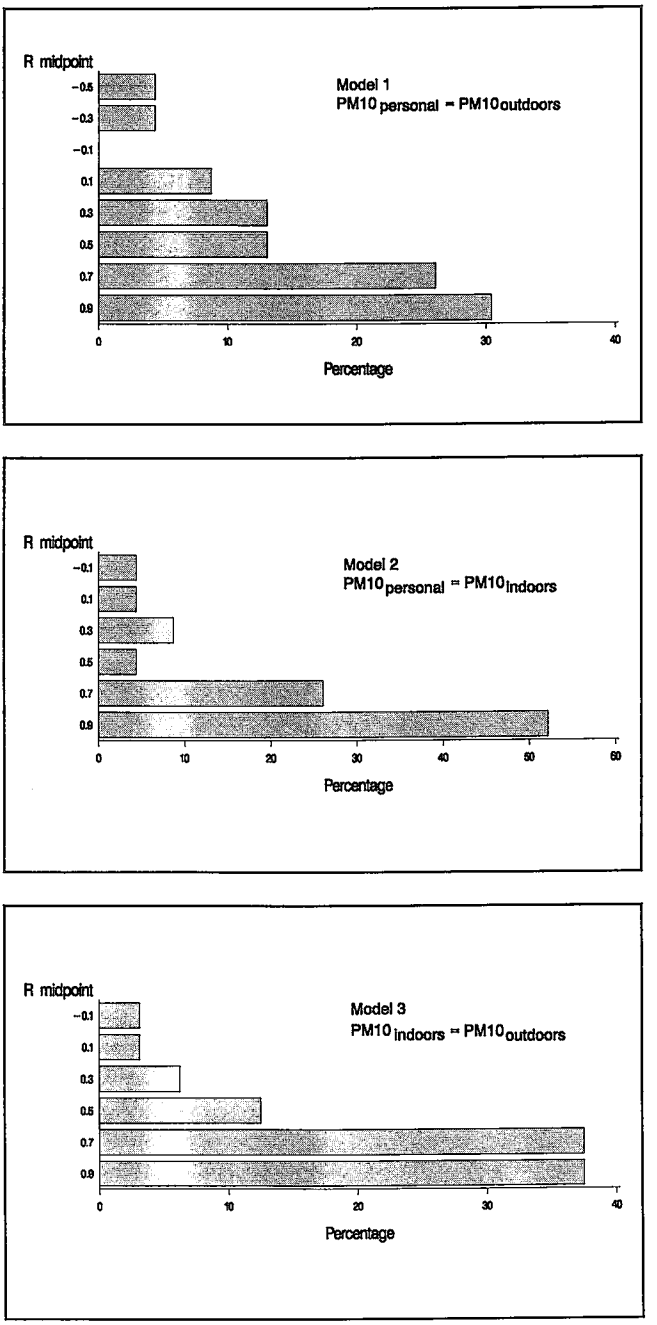

Figure 2. Distribution of individual Pearson's correlation coefficients from 50- to 70-year- old adults, after excluding days with exposure to ETS (n = 23 for models 1 and 2; n = 32 for model 3)

The median value of 1,000 cross-sectional Pearson's correlation coefficients was 0.34 (range -0.09 to 0.67) when selecting from all observations, and 0.50 (range -0.07 to 0.83) when only days with no exposure to ETS were selected.

Difference between personal, indoor and outdoor PM10 concentrations

In table 1 we showed that personal exposures exceeded indoor and outdoor concentrations. The mean difference between personal and outdoor concentrations was 20 $\mu g/m^3$. Indoor concentrations were on average 7 $\mu g/m^3$ lower than the corresponding outdoor concentrations, ruling out the concentrations indoor as a possible explanation for the excess personal exposures. Furthermore, the higher personal exposures can not be explained by the use of different samplers for personal and outdoor measurements, because outdoor concentrations measured with the personal impactor did not significantly differ from concentrations measured with the outdoor sampler[15].

Results of the analyses of the relationship between the difference between personal and outdoor concentrations and several personal characteristics and activities are presented in table 4. Exposure to ETS (both at home and elsewhere), living along a busy road and time spent in a vehicle significantly contributed to the difference between personal and ambient concentrations. Cleaning activities, cooking, time spent outdoors, sex and ventilation did not have a significant effect. The intercept of the model is 4 $\mu g/m^3$ and does not significantly deviate from zero.

Results of the regression analyses of the difference between indoor and outdoor concentrations are presented in table 5. Smoking in the living room and cooking in a kitchen which was in open connection with the living room significantly contributed to the difference between indoor and outdoor concentrations. In contrast to what we found for personal exposures, indoor concentrations were not higher in the living room of subjects who lived along a busy road when compared to concentrations in the other living rooms.

Table 4. Multiple regression analysis of the relationship between the difference between personal and outdoor PM10 (μg/m³) and several other variables (n = 256)

	Parameter Estimate	Se	95% CI†	Mean of the variable
Intercept	4.35	5.96	-7.76 to 16.46	
# cigarettes smoked in the living room	2.33**	0.70	0.94 to 3.72	0.56‡
# hours spent in the presence of smokers	5.70**	1.38	2.98 to 8.43	0.57§
Living along a busy road (yes/no)	22.73**	5.36	11.84 to 33.62	0.20
Time spent in a vehicle (hours)	5.42*	2.73	0.05 to 10.80	0.29
Cooking (yes/no)	4.82	3.89	-2.86 to 12.50	0.81
Cleaning activities (yes/no)	2.16	3.06	-3.87 to 8.19	0.58
Time spent outdoors (hours)	-1.19	1.33	-3.81 to 1.43	1.29
Sex (♀ = 0; ♂ = 1)	3.80	4.37	-5.08 to 12.67	0.50
Living room window opened (yes/no)	-1.60	3.61	-8.71 to 5.51	0.38
Slept with bedroom window opened (yes/no)	1.40	3.76	-6.01 to 8.80	0.61

* $p < 0.05$

** $p < 0.01$

† *Confidence interval*

‡ *Smoking in the living room was reported 26 times*

§ *Exposure to ETS elsewhere was reported 64 times*

Table 5. Multiple regression analysis of the relationship between the difference between indoor and outdoor PM10 (μg/m³) and several other variables (n = 241)

	Parameter Estimate	Se	95% CI †	Mean of the variable
Intercept	-12.48**	3.32	-19.23 to -5.74	
# cigarettes smoked in the living room	2.33**	0.51	1.32 to 3.34	0.59‡
Cooking, kitchen in living room (yes/no)	6.95*	3.94	-0.81 to 14.71	0.20
Cooking, kitchen elsewhere (yes/no)	0.60	3.04	-5.40 to 6.59	0.62
Cleaning activities (yes/no)	2.97	2.31	-1.59 to 7.52	0.58
Living along a busy road (yes/no)	-2.12	3.48	-9.19 to 4.95	0.17
Living room window opened (yes/no)	2.19	2.19	-2.67 to 7.04	0.39

* $p < 0.10$

** $p < 0.001$

† *confidence interval*

‡ *Smoking in the living room was reported 23 times*

Discussion

Correlation between personal, indoor and outdoor PM10 concentrations

In this study we found a reasonably high correlation between personal and outdoor PM10 concentrations, within subjects, over time, despite a relatively small range in outdoor concentrations. For non-ETS exposed subjects, daily variations in ambient PM10 concentrations accounted for about 50 percent of the variation in personal exposures. The correlation between personal and indoor, and indoor and outdoor concentrations was even better. Correlations within subjects over time were higher than the cross-sectional correlation.

Some recent studies have also shown higher within subject correlations than cross-sectional correlations[11,19]. In a similar study among 45 children aged 10 to 12 years, we found a median Pearson's R between personal and outdoor PM10 concentrations of 0.63 compared to a cross-sectional correlation of 0.28[19]. In the PTEAM pilot study, repeated measurements of PM10 were conducted in nine households (two persons in each household). Cross-sectionally, personal exposures were uncorrelated with outdoor concentrations but for the 10 subjects (five homes) with 6 to 8 individual measurements, individual correlations ranged from -0.17 to 0.79, with a median value of 0.26[11]. In the Total Human Environmental Exposure Study (THEES), Buckley *et al.*[13] calculated the correlation within subjects, using 9 to 14 personal PM10 measurements from 13 non-smoking adults. Individual coefficients of the correlation between personal and ambient concentrations ranged from 0.14 to 0.90 with a median value of 0.53. Wallace[11] presented both the cross-sectional and the within subject correlations using data from 14 subjects in the THEES study. The cross-sectional correlation between personal and outdoor concentrations was 0.52 (n = 181), whereas the median of the individual correlations was 0.68 (range 0.14 to 0.91). Lioy *et al.*[20] reported the indoor-outdoor correlations of eight homes in the THEES study. The cross-sectional correlation (n = 101) was 0.67, compared with a median individual correlation coefficient of 0.88 (range 0.60 to 0.98)[11].

After excluding days with exposure to ETS, the correlation coefficients increased. In the similar study on childhood exposure to PM10, excluding days that children with non-smoking parents were exposed to ETS increased the correlation from a median R of 0.63 to a median R of 0.73. In the THEES study[13] using activity data improved the personal estimates for all individuals, to

correlation coefficients ranging from 0.58 to 0.999 with a median value of 0.93. Exposure to ETS was one of the activity variables that contributed to the improvement of the individual correlations, together with house-cleaning activities, cooking and use of unvented kerosene space heaters. Correlations after accounting for exposure to ETS alone were not described.

The median slope was about 0.5 for model 1 (personal-outdoor) and model 3 (indoor-outdoor) and close to 1 for model 2 (personal-indoor). These values are comparable to those found in the THEES and PTEAM study[11].

Individual correlation coefficients ranged from moderately negative to strongly positive values. Because of the limited number of observations per subject used to calculate the individual correlation coefficients, however, precision of individual estimates is low. Most value should therefore be put on the population median instead of individual values.

It has been argued that the low correlation between personal and outdoor exposure to particles makes associations between day-to-day variations in outdoor air pollution and health effects implausible. The significant correlation between outdoor and personal exposure found in this study documents, however, that short-term increases in outdoor air pollution are reflected in increased personal exposures. This finding provides support for using fixed site measurements as a measure of exposure to PM10 in time series studies linking the day-to-day variation in PM10 to the day-to-day variation in health endpoints.

Difference between personal, indoor and outdoor PM10 concentrations

Personal exposures considerably exceeded outdoor and indoor concentrations. The major part of the difference between personal and outdoor concentrations, however, could be attributed to exposure to ETS, living along a busy road and time spent in a vehicle. Indoor concentrations in the living room were lower than outdoor concentrations, and were increased in case of smoking and cooking in a kitchen in open connection to the living room.

An important part of the difference between personal and outdoor concentrations was attributed to exposure to ETS. Although all participants were non-smokers with no smokers in their households, 21 subjects reported exposure to ETS on at least one of the days of personal measurements. The majority of the exposure to ETS occurred outside their own home-environment; only seven subjects reported exposure to ETS in their own living room. The estimated contribution of one cigarette to the 24 hour average personal and

indoor PM10 concentration was 2.3 $\mu g/m^3$, which is slightly higher than the range of 1 to 2 $\mu g/m^3$ that was recently suggested for PM2.5 by Wallace[11].

Subjects who lived along a busy road had higher personal exposures than subjects who did not live along a busy road. The estimated difference, adjusted for other factors such as exposure to ETS, was 23 $\mu g/m^3$. Indoor concentrations, however, were not higher in homes along busy streets. One possible explanation for this inconsistency might be that subjects who live along busy roads are exposed to higher PM10 concentrations when they go outdoors. Janssen *et al.*[21] found significantly higher daytime PM10 concentrations on the pavement of two busy roads compared with simultaneously measured background concentrations. The mean differences, however, were small: 7 $\mu g/m^3$ for the road in a town (traffic intensity 8,900 vehicles per 24 hours) and 13 $\mu g/m^3$ for the road in a medium-sized city (traffic intensity 15,000 vehicles per 24 hours). Bevan *et al.*[22] measured exposure to RSP while commuting by bicycle during peak traffic hours. The mean RSP concentration when cycling through a typical "urban" environment was 139 $\mu g/m^3$, compared with 120 $\mu g/m^3$ when cycling through a "suburban" area. Another aspect might be that we placed the equipment in the main living area, not necessarily being the road side of the house. Fischer *et al.*[23] and Oldenwening *et al.*[24] measured indoor and outdoor 24-hour averaged PM10 and PM2.5 concentrations in 30 houses in Amsterdam. Only homes with the living room on the roadside were selected. The mean indoor PM10 concentrations along busy roads were about 9 $\mu g/m^3$ higher than the mean concentration in the houses that were situated on more quiet streets. Though these studies confirm the plausibility of higher particle concentrations near busy roads, our estimated difference of 23 $\mu g/m^3$ seems rather large. Furthermore, the inconsistency of significant higher personal exposures for subjects living along busy roads, but no difference in indoor concentrations can not readily be explained. Possibly some other characteristics associated with living along a busy road are responsible for the effect.

Time spent in a vehicle also significantly contributed to the difference between personal and outdoor concentrations. The estimated contribution was 5.4 $\mu g/m^3$ per hour spent in a vehicle. To cause such an increase in the 24-hour averaged personal concentration, the PM10-concentration in the vehicle must have been about 130 $\mu g/m^3$ (24 hours x 5.4 $\mu g/m^3$ per hour) higher than the outdoor concentration. Although several studies have been conducted on the exposure of car drivers to gaseous traffic related air pollutants[25-27], limited

information is available about particle concentrations inside vehicles. Morandi *et al.*[28] measured personal RSP concentrations of 30 subjects for 12 hours, using a portable piezobalance-type respirable mass monitor with 5-minute integration times. The mean RSP concentration inside vehicles was 35 $\mu g/m^3$, significantly higher than the mean outdoor concentrations of 22 $\mu g/m^3$, but suggesting smaller differences than the difference necessary to explain our estimated contribution of 5.4 $\mu g/m^3$ per hour. However, the results are not directly comparable because of the difference in the particle sizes measured (RSP versus PM10). Possibly resuspension of the coarse part of PM10 particles, caused by the presence of persons in the small volume of a car, is responsible for (part of) the difference.

Cooking in a kitchen with an open connection to the living room increased the indoor PM10 concentrations. The influence of cooking on personal exposures was lower and not significant. Cleaning activities did not have a significant effect on personal or indoor concentrations. In addition to exposure to ETS, several studies identified cooking as a second important source of particles[11]. In the PTEAM study, Ozkaynak *et al.*[29] found that cooking added about 12 $\mu g/m^3$ and 26 $\mu g/m^3$ to nighttime and daytime indoor PM10-concentrations, respectively. Other household activities such as vacuuming and dusting appeared to make smaller contributions to indoor particle levels. Morandi *et al.*[28] also found higher RSP concentrations in the presence of active cooking (mean 27 $\mu g/m^3$) than in the absence of cooking emissions (mean 20 $\mu g/m^3$). Buckley *et al.*[13] reported that house-cleaning and cooking were important activity variables in improving the correlation between personal and outdoor PM10-concentrations. Quantitative information about the contribution of these activities, however, was not provided.

Excess personal exposures compared to indoor or outdoor concentrations have been found in most personal exposure studies[11], with the exception of some studies among disabled or retired persons[30] and patients with severe chronic obstructive pulmonary disease[31]. Resuspension of coarse particles by personal activities and proximity to particle-generating sources have been suggested as causes of this so-called personal cloud[11]. For the older adults studied in this study, the major part of the difference between personal and outdoor PM10 concentrations could be attributed to exposure to ETS, living along a busy road and time spent in a vehicle.

In conclusion, this study has shown that personal PM10 concentrations are reasonably well correlated with ambient PM10 concentrations, within subjects, over time. This finding provides support for using fixed site measurements as a measure of exposure to PM10 in time series studies linking the day-to-day variation in PM10 to the day-to-day variation in health endpoints.

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4. Childhood exposure to PM10: relation between personal, classroom and outdoor concentrations*

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Abstract

To investigate the validity of outdoor PM10 concentrations as a measure of exposure in time series studies, the association between personal and outdoor concentrations, within children, over time was investigated. Four to eight repeated measurements of personal and outdoor PM10 concentrations were conducted among children, aged 10 to 12 years, from four schools in Wageningen and Amsterdam, the Netherlands. Repeated PM10 measurements in the classrooms were conducted in three of the schools. Averaging time was 24 hours for the personal and outdoor measurements, and 8 hour (daytime) and 24 hour for the classroom measurements. For each child separately, personal exposures were related to outdoor concentrations in a regression analysis. The distribution of the individual correlation and regression coefficients was investigated. Information about factors that might influence personal exposures was obtained by questionnaire. Median Pearson's R between personal and outdoor concentrations was 0.63 for children with non-smoking parents and 0.59 for children with smoking parents. For children with non-smoking parents, excluding days with exposure to ETS improved the correlation to a median R of 0.73. The mean personal PM10 concentration was 105 $\mu g/m^3$; on average 67 $\mu g/m^3$ higher than the corresponding outdoor concentrations. The main part of this difference could be attributed to exposure to ETS, to high PM10 concentrations in the classrooms, and to (indoor) physical activity. The results show a reasonably high correlation between repeated personal and outdoor PM10 measurements within children, providing support for the use of fixed site measurements as a measure of exposure to PM10 in epidemiological time series studies. The large differences between personal and outdoor PM10 concentrations are probably a result of a child's proximity to particle-generating sources and particles resuspended by personal activities.

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Introduction

Several recent studies have documented associations between the day-to-day variation of particulate matter air pollution and acute health effects on children, including increased respiratory symptoms and decreased lung function[1-5]. In these studies, exposure assessment was based on fixed site measurements in ambient air. Measurements of personal exposure are considered to be a more accurate estimate of the subject's true exposure[6]. Children's personal exposures to particles have rarely been studied. Studies among adults suggest that outdoor particle mass concentrations correlate poorly with personal exposures[7]. If the variation in outdoor levels of particulate matter is not tightly linked to variation in personal exposures, use of outdoor concentrations as a surrogate for personal exposures would tend to misclassify personal exposures and exposure-response relationships could be attenuated[8]. However, in most personal exposure studies the correlation between personal and outdoor concentrations was calculated cross-sectionally: personal exposure data were collected from a group of subjects by measuring different subsets of subjects on different days (=different ambient concentrations) and measuring each subject once or only a limited number of times. Next, one correlation coefficient was calculated, using all measurements from all subjects and days. This correlation is influenced by the variation in personal exposure between subjects. As time series studies relate day-to-day variations in outdoor concentrations to day-to-day variations of health endpoints, the correlation between personal and ambient concentrations within a person, over time, is more relevant than the variation between persons. This correlation may be better because factors that can cause variation between subjects, such as exposure to environmental tobacco smoke (ETS), are less variable in time within subjects, and therefore mainly cause variation between subjects. At present, only limited information is available about the within-subject correlation between personal and outdoor PM10 concentrations[9,10].

To investigate the validity of outdoor concentrations as a measure of exposure to PM10 in time series studies, we conducted a personal exposure study in which repeated measurements of personal and outdoor PM10 were conducted to allow calculation of the correlation between outdoor and personal measurements of PM10, within subjects, over time. This paper describes the relationship between personal and outdoor PM10 concentrations in a group of 10- to 12-year-old children.

Methods

Study design

Children, aged 10 to 12, were recruited through schools; two in Wageningen, a non-industrial town of about 35,000 inhabitants, and two in Amsterdam, the capital of The Netherlands. All children in one class were asked to participate by means of a presentation in their classroom, which included a demonstration of the sampling equipment. The children received a written description and informed consent form for their parents. Out of 57 children in Wageningen 33 (58%) and out of 56 children in Amsterdam 15 (27%) participated. Of those 48 children 45 successfully completed the study.

Measurements took place in two periods: from 16 February to 19 April 1994 (one school in Wageningen) and from 11 January to 18 May 1995 (three schools). Seven to eight measurements per child were planned. 24-hour averaged measurements of personal PM10 were conducted on weekdays only, spaced approximately one week apart. Samplers were distributed and collected at school, except for the first measurement when samplers were distributed at the children's homes to provide individual instruction to the children in the presence of a parent. Children were instructed to wear the sampler whenever possible. At night the sampler was placed near the bed. Outdoor PM10 measurements were conducted at fixed sites (see below). PM10 measurements in classrooms were added to the study in 1995 (three schools).

Information on general characteristics such as parental smoking was collected by questionnaire. In addition, parents were asked to fill out a questionnaire about each day of personal measurements, including questions on exposure to ETS, time spent in several micro-environments, cleaning and cooking activities conducted by or in the presence of the child. After each day of measurements these questionnaires were collected at the children's homes and checked for completeness and accuracy of the answers. Exposure to ETS was assessed by means of the following questions:

1) Has anybody smoked in your living room during the measurements? yes / no
2) Has your child been in a room, other than your own living room, where people smoked? yes / no

In 1995 a question on physical activity was added to the questionnaire:

Has your child conducted any activities during which he/she was physically active? yes / no

If yes, what kind of activities? _____

Examples of activities were given to clarify this question.

Sampling methods

Personal measurements were conducted using a personal impactor described by Buckley *et al.*[9] using 25 mm diameter 3 μm pore size Gelman Teflon filters and a flow-controlled battery operated pump (Gillian, model Gil-air 5). Details about the sampling method and quality issues are described elsewhere[11].

Outdoor PM10 measurements were conducted using the personal sampler at a fixed site. In Amsterdam this site was located in a park in the city centre, about 150 metres away from the nearest road; in Wageningen the equipment was placed in a meadow on the outskirts of the town about 500 metres away from the nearest road. Both sites were away from local particle sources such as unpaved roads, construction work or industrial sources. Measurements were conducted at a height of 1.5 m. Co-located operation of the personal sampler with a Harvard Impactor and a Sierra Anderson sampler in Amsterdam showed highly correlated outdoor concentrations (R 0.91 - 0.95) and no significant differences in concentrations obtained with the different methods[11].

In 1995, PM10 measurements in classrooms were conducted with a Harvard Impactor (ADE Inc, Harrison, Maine, USA)[12]. A flow controlled pump (ADE Inc, model SP-280E) and Anderson 37 mm 2 μm pore size Teflon filters were used. Measurements were conducted at a height of 1.5 m, away from the door and the blackboard. Two averaging times were used: 24 hour measurements at the same time as the personal measurements ($\pm$15.00-15.00) and 8 hour measurements at the time the children were at school ($\pm$8.00-16.00).

For the personal and classroom measurements, flows were measured at the beginning and end of each sampling period with calibrated rotameters, and elapsed time indicators were used to calculate the sampled volumes. For the outdoor measurements sampled volumes were determined with calibrated dry gas metres. Measurements that had lasted for less than 20 hours were excluded.

In 1994, filters were weighed using an analytical balance with 10 μg reading. In 1995 a micro-balance was used to weigh the personal and outdoor filters. Classroom filters were weighed on the analytical balance. All filters were weighed

twice after equilibrating at 20 °C and 44% relative humidity for 24 hours in a desiccator. Mean field blank weight changes were subtracted from all sample weights. Detection limits, defined as 3 times the standard deviation in field blanks divided by the sampled volume, of personal and outdoor measurements were 10.8 $\mu g/m^3$ in 1994 and 8.6 $\mu g/m^3$ in 1995. The detection limit of the classroom measurements was 3.7 $\mu g/m^3$ for the 24 hour measurements and 11.1 $\mu g/m^3$ for the 8 hour average. All measurements were above the detection limit.

Data Analysis

Correlation between personal and outdoor concentrations

The correlation between personal and outdoor PM10 concentrations was assessed by means of individual regression analysis, with the following model:

$PM10_{personal,i\,t} = \alpha_i + ß_i \times PM10_{outdoors,\,t}$ Where i = child i and t = day t

The distribution of the individual regression results was investigated. Medians are presented because not all regression results were normally distributed for all models. 95% confidence intervals were calculated using a non-parametric method published by Campbell and Gardner[13].

All children were non-smokers. No selection on parental smoking was made, however. Furthermore, children with non-smoking parents could be exposed to ETS elsewhere or at home in the case of a visitor who smoked. The influence of exposure to ETS on the relationship between personal and outdoor concentrations was investigated by stratifying for parental smoking and by excluding days that children with non-smoking parents were exposed to ETS.

The influence of time spent outdoors on the relationship between personal and outdoor PM10 was assessed by adding an interaction term 'much time spent outdoors x outdoor concentration' to the regression model. The variable 'much time spent outdoors' was assigned one for measurement days that a child had spent more time outdoors than the median amount of time spent outdoors and zero for the other days.

The influence of PM10 exposure in the classroom on the relationship between personal and outdoor PM10 was assessed by regressing the personal exposures against a time weighted concentration, C_{tw}:

$C_{tw} = (C_{classroom,\ 8\ hours} \times 6\ hours + C_{outdoors,\ 24\ hours} \times 18\ hours)/24\ hours$

with: $C_{classroom,\ 8\ hours}$ = 8 hour average concentration in the classroom
$C_{outdoors}$ = concentrations outdoors (24 hour average)
6 hours = number of hours spent at school
18 hours = 24-6

Regression analyses with the outdoor and classroom concentrations as two separate independent variables was not conducted because for one schools daytime classroom concentrations were highly correlated with outdoor concentrations (R 0.91).

For comparison purposes, we calculated what the correlation would have been, if it had been calculated cross-sectionally. In this analysis, we randomly selected one measurement per subject and next calculated the cross-sectional correlation between personal and outdoor concentrations. This procedure was repeated 1,000 times and the average of those 1,000 correlation coefficients was calculated to get a more reliable estimate of the cross-sectional correlation.

Difference between personal and ambient concentrations

The questionnaire data were used to examine to what extent differences between personal and ambient concentrations could be explained by certain activities, such as exposure to ETS. The difference between personal exposures and time weighted concentrations was used as the dependent variable in a regression analysis. The SAS (Statistical Analysis System) procedure "PROC MIXED" was used to adjust regression results for correlations between repeated measurements. A random intercept model was used. Different questions on cleaning activities (dusting, vacuum cleaning, sweeping and cleaning a pet's cage) were combined into one variable 'cleaning activities'. Physical activities were divided into four categories: 'active indoors, pump nearby or carried', 'active outdoors, pump carried', 'active outdoors, pump nearby' and a fourth variable 'active, pump elsewhere' which included activities during which the pump had not been at the same site as the child. 'Active indoors, pump nearby or carried' was not divided into two variables because for all but five occasions that indoor physical activities were reported, subjects also reported not to have carried the pump.

Results

Population

A total of 45 children, 21 boys and 24 girls, participated. 13 children lived in Amsterdam and 32 in Wageningen. 20 Children had a parent who smoked. Of those children 18 were exposed to ETS on all measurement days; two children had one day without exposure to ETS. On days of personal measurements, children spent on average 2.7 hours outdoors, 14.9 hours at home and 5.7 hours at school.

Concentration levels

From the 45 participating children 334 personal measurements were conducted, of which 33 samples (9.6%) were lost, mostly due to pump failure[11]. The distributions of the individual averages of personal and outdoor PM10 are presented in table 1. Personal exposures were on average 67 $\mu g/m^3$ higher than outdoor concentrations. The average coefficients of the variation (CV) in personal, outdoor and the difference between personal and outdoor concentrations were 22.6%, 56.0% and 37.6% respectively. Table 2 shows the results of the PM10 measurements in the classrooms. In all schools, PM10-concentrations during school hours were much higher than during non-school hours. Classroom concentrations, both 24 hour averaged and 8 hour averaged, were significantly higher than outdoor concentrations. Striking is that in the school with the lowest classroom concentrations (Amsterdam, school 2), the lowest personal concentrations were measured. Table 3 shows the distribution of the time weighted concentrations. Personal exposures were on average 43 $\mu g/m^3$ higher than the time weighted concentrations. This will be discussed in more detail later.

Table 1. Distribution of individual averages of personal and ambient PM10

			Personal ($\mu g/m^3$)			Ambient ($\mu g/m^3$)			Difference ($\mu g/m^3$)		
	n	(#*)	Mean	Sd	Range	Mean	Sd	Range	Mean	Sd	Range
Wageningen 1994	15	(89)	111.0	23.4	83.7 to 167.0	43.0	7.2	24.5 to 55.8	68.1	27.3	40.6 to 142.4
Amsterdam 1995, school 1	6	(41)	105.3	16.6	76.1 to 121.8	39.2	1.1	37.5 to 40.8	66.1	17.1	35.3 to 81.7
Amsterdam 1995, school 2	7	(48)	88.8	31.6	56.9 to 140.2	36.5	2.6	32.7 to 39.1	52.3	31.4	22.7 to 106.6
Wageningen 1995	17	(123)	106.8	34.2	71.3 to 195.4	35.0	2.3	29.7 to 39.1	71.8	34.7	35.4 to 160.1
Total	45	(301)	105.2	28.7	56.9 to 195.4	38.5	5.6	24.5 to 55.8	66.8	29.8	22.7 to 160.1

* *Number of measurements*

Table 2. PM10 concentrations in classrooms (μg/m³)

	Amsterdam 1995, school 1 (n = 15)			Amsterdam 1995, school 2 (n = 15)			Wageningen 1995 (n = 11)		
	Mean	Sd	Range	Mean	Sd	Range	Mean	Sd	Range
8 hour average	157.0	38.8	96.2 to 234.1	80.8	18.7	57.1 to 127.0	134.1	42.1	66.3 to 198.6
24 hour average	74.4	19.6	32.1 to 108.2	45.9	13.9	30.7 to 79.5	63.1	20.7	37.8 to 105.6
Outdoor	34.0	14.0	14.7 to 75.2	34.5	13.8	14.7 to 75.2	32.0	14.4	16.6 to 71.5
Estimated 16 hour†	37.0	17.4	-1.1 to 58.7	30.0	14.8	12.9 to 64.1	33.7	21.8	-0.7 to 80.3
Difference 8-24‡	82.6***	25.1	34.1 to 126.0	34.9***	11.9	15.7 to 64.9	71.0***	29.0	27.9 to 112.4
Difference 8-24§	123.0***	41.8	57.2 to 200.8	46.4***	13.1	28.6 to 76.5	102.1***	36.3	49.7 to 149.8
Difference 24-outdoor¶	40.4***	21.3	1.0 to 74.8	11.5**	11.9	-2.2 to 40.5	31.1***	16.8	11.6 to 72.4

† *Estimated classroom concentration during non-school hours:*
Estimated 16 hours $= (C_{24\ hours} \times t_{24\ hours} - C_{8\ hours} \times t_{8\ hours})/(t_{24\ hours} - t_{8\ hours})$

‡ *Difference between 8 hour averaged and 24 hour averaged concentrations*

§ *Difference between 8 hour averaged and outdoor concentrations*

¶ *Difference between 24 hour averaged and outdoor concentrations*

** *t-test mean = 0; $p < 0.01$*

*** *t-test mean = 0; $p < 0.001$*

Table 3. Distribution of individual averages of personal and time weighted PM10 concentrations (μg/m³)

	All children (n=30)			Children with non-smoking parents (n=16)			Children with smoking parents (n=14)		
	Mean	Sd	Range	Mean	Sd	Range	Mean	Sd	Range
Personal	102.3	30.9	56.9 to 195.4	84.0	17.3	56.9 to 126.4	123.3	30.0	80.1 to 195.4
C_{tw}*	58.9	7.5	42.5 to 67.2	58.9	8.0	42.6 to 67.2	58.9	7.3	42.5 to 65.8
Difference	43.4	30.8	8.7 to 134.2	25.1	14.8	8.7 to 64.4	64.4	31.2	31.8 to 134.2

* *time weighted average of outdoor and classroom PM10 concentrations*

Correlation between personal and outdoor concentrations

Results from the individual regression analyses for the relation between personal and outdoor concentrations are presented in table 4 and figure 1. Median Pearson's R was 0.63. Median R and slope were similar for children with non-smoking parents and children with smoking parents. The median intercept was higher for children with smoking parents. Adding information about time spent outdoors did not improve the correlations: the median slope of the interaction term 'much time spent outdoors x outdoor concentration' was -0.02 and highly non-significant ($p = 0.82$). No consistent differences between Wageningen and Amsterdam were found, both in correlations and in slopes. The average range (maximum minus minimum) in outdoor concentrations on days of personal sampling was 63 $\mu g/m^3$ (sd 29; range 13 to 105 $\mu g/m^3$). Excluding children with a range smaller than 25 $\mu g/m^3$ (four children), resulted in exclusion of the two highest slopes (>2.5), but did not significantly change the medians.

Table 5 and figure 2 show the results from the regression analyses with the time weighted concentrations instead of the outdoor concentrations. Because measurements in the classrooms were not conducted in 1994, only 30 children are included in table 5. For these 30 children, median Pearson's R increased from 0.58 to 0.67 for all children, from 0.61 to 0.70 for children with non-smoking parents and from 0.47 to 0.60 for children with smoking parents.

Table 6 and figure 3 show the results after excluding days that children with non-smoking parents were exposed to ETS. Of the 25 children included in table 4, 10 were occasionally exposed to ETS and therefore had different regression results in table 6 compared with table 4. For these 10 children, median Pearson's R increased from 0.51 to 0.73. For the correlation between personal and time-weighted concentrations, eight children had different regression results in table 6 compared with table 5 and for these 8 children median R increased from 0.65 to 0.79.

The mean value of 1,000 cross-sectional Pearson's correlation coefficients between personal and outdoor concentrations was 0.28 (sd 0.12; range -0.11 to 0.60) for all children, 0.45 (sd 0.16; range -0.23 to 0.82) for children with non-smoking parents and 0.20 (sd 0.19; range -0.46 to 0.82) for children with smoking parents.

Table 4. Distribution of individual regression results, regression of $PM10_{personal}$ (Y-variable) on $PM10_{outdoors}$ (X-variable)

	All children (n = 45)		Children with non-smoking parents (n = 25)		Children with smoking parents (n = 20)	
	Median	95% CI*	Median	95% CI*	Median	95% CI*
Intercept	75.4	68.4 to 86.9	69.5	53.2 to 75.5	97.3	75.9 to 114.0
Slope	0.57	0.43 to 0.75	0.57	0.40 to 0.77	0.60	0.28 to 1.00
Pearson's R	0.63	0.50 to 0.72	0.63	0.50 to 0.80	0.59	0.36 to 0.80

* *95% confidence interval*

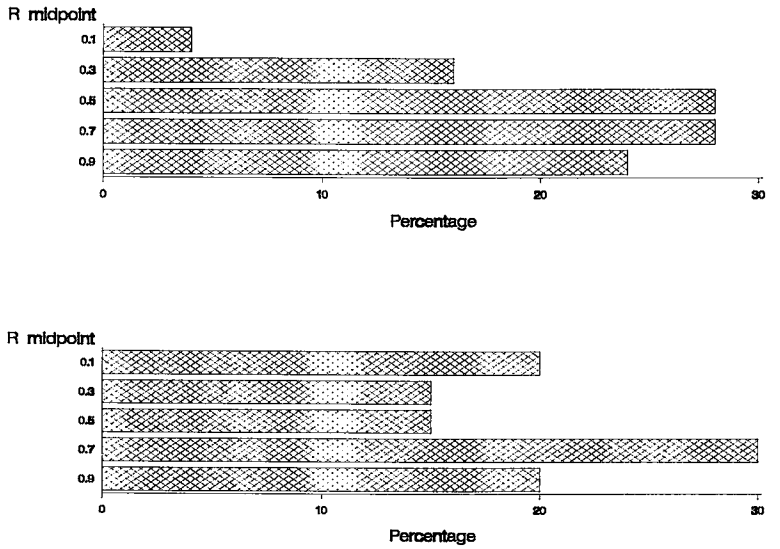

Figure 1. Distribution of individual Pearson's correlation coefficients between personal and outdoor concentrations for children with non-smoking parents (upper; n = 25) and smoking parents (lower; n = 20)

Table 5. Distribution of individual regression results, regression of $PM10_{personal}$ (Y-variable) on $PM10_{tw\text{-}model}$ (X-variable)

	All children (n = 30)		Children with non-smoking parents (n = 16)		Children with smoking parents (n = 14)	
	Median	95% CI*	Median	95% CI*	Median	95% CI*
Intercept	57.2	40.6 to 70.6	42.5	23.2 to 58.4	76.9	53.9 to 147.5
Slope	0.67	0.53 to 0.76	0.65	0.50 to 0.99	0.70	0.03 to 0.81
Pearson's R	0.67	0.52 to 0.81	0.70	0.59 to 0.83	0.60	0.02 to 0.81

* *95% confidence interval*

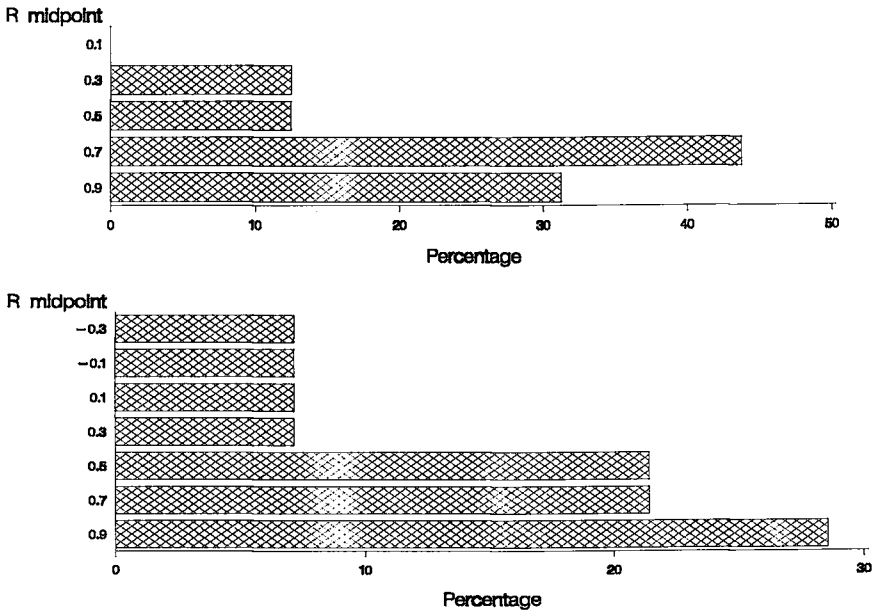

Figure 2. Distribution of individual Pearson's correlation coefficients between personal and time weighted concentrations for children with non-smoking parents (upper; n = 16) and smoking parents (lower; n = 14)

*Table 6. Distribution of individual regression results for children with non-smoking parents, after excluding days with exposure to ETS**

	$PM10_{personal}$ = $PM10_{outdoors}$ (n=25)		$PM10_{personal}$ = $PM10_{tw\text{-}model}$ (n=16)	
	Median	95% CI*	Median	95% CI*
Intercept	61.4	43.6 to 76.1	37.7	22.9 to 50.8
Slope	0.57	0.41 to 0.86	0.72	0.50 to 0.99
Pearson's R	0.73	0.56 to 0.83	0.76	0.67 to 0.89

* *95% confidence interval*

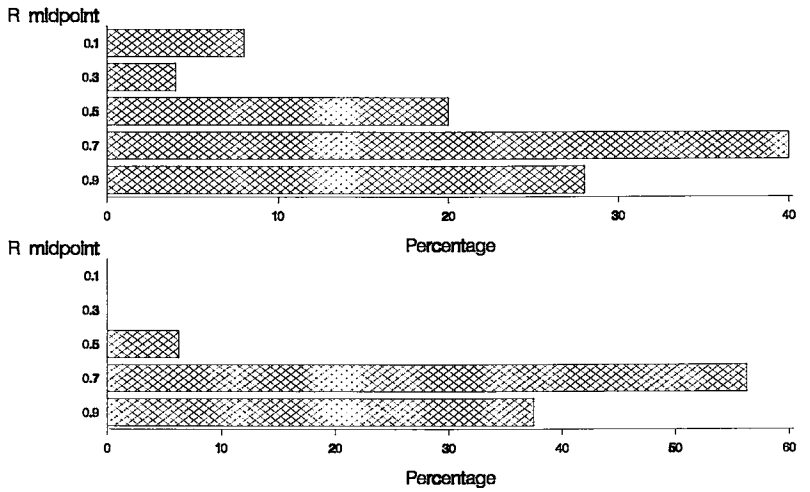

Figure 3. Distribution of individual Pearson's correlation coefficients between personal and outdoor concentrations (upper; n=25) and between personal and time weighted concentrations (lower; n=20) for children with non-smoking parents, after excluding days with exposure to ETS

Difference between personal and ambient concentrations

In table 1 and 3 it was shown that personal exposures were on average 67 $\mu g/m^3$ higher than outdoor concentrations and 43 $\mu g/m^3$ higher than time weighted concentrations. The mean difference between personal and time weighted

concentrations was 25 $\mu g/m^3$ for children with non-smoking parents and 64 $\mu g/m^3$ for children with smoking parents. The average difference between personal and time weighted concentrations per school ranged from 39 to 45 $\mu g/m^3$; a much smaller range than the range presented in table 1 (52 to 72 $\mu g/m^3$).

Table 7 shows the results from the analyses of the relationship between the difference between personal and time weighted concentrations and several personal characteristics or activities. Exposure to ETS and physical activity significantly contributed to the difference between measured personal exposures and the time-weighted model predictions. Dividing the physical activities into categories only showed a significant influence of 'active indoors'. The other activity categories did not have a significant effect. The parameter estimate for 'active indoors' was 12.9 $\mu g/m^3$ (SE 3.7). Time spent outdoors, both as a continuous variable and as a binary variable, did not consistently influence personal exposures and is therefore not included in the model presented in table 7. The intercept of the model is 6.6 $\mu g/m^3$ and not significantly different from zero.

Table 7. Multiple regression analysis of the relationship between the difference between personal and time weighted concentrations and several other variables (n = 208)

	Parameter estimate	se	95% CI[†]	Mean of variable
Intercept	6.64	8.25	-9.53 to 22.81	
Smoking parent(s) (yes/no)	41.28**	8.64	24.34 to 58.22	0.45
Exposure to ETS[‡] (yes/no)	12.29*	5.13	2.24 to 22.33	0.11
Physical activity (yes/no)	11.61**	3.70	4.36 to 18.86	0.63
Sex (girl = 0; boy = 1)	10.80	8.88	-6.60 to 28.21	0.62
Time spent in a vehicle (yes/no)	3.42	5.49	-7.34 to 14.19	0.15
Cooking[§] (yes/no)	1.24	3.54	-5.69 to 8.17	0.59
Cleaning activities[§] (yes/no)	3.84	4.34	-4.66 to 12.34	0.30
Living room window opened (yes/no)	0.98	4.82	-8.46 to 10.42	0.20
Slept with bedroom window opened (y/n)	0.45	4.70	-8.77 to 9.66	0.33

* $p < 0.05$

** $p < 0.01$

† *confidence interval*

‡ *other than caused by parental smoking*

§ *conducted by or in the presence of the child*

Discussion

Correlation between personal and outdoor concentrations

This study has shown that personal PM10 concentrations are reasonably well correlated with ambient PM10 concentrations, within children, over time. The median of the individual correlation coefficients was 0.63 for children with non-smoking parents and 0.59 for children exposed to parental smoking. The estimated cross-sectional correlation coefficients were considerably lower: 0.47 and 0.20 for children with non-smoking and smoking parents, respectively.

Exposure to ETS significantly increased personal PM10 exposures. For children with smoking parents, smoking in the living room was reported on all but two occasions. The median R of 0.59 found for children with smoking parents shows that, despite the significant influence of parental smoking on the level of exposure, personal PM10 exposures of children exposed to ETS on a day-to-day basis are still reasonably well correlated with outdoor concentrations. Excluding days that children with non-smoking parents were exposed to ETS improved the correlation to a median R of 0.73.

PM10 concentrations in the classrooms were a second important cause of excess personal exposures. Correlating the personal exposure with concentrations calculated with a time weighted model that accounted for the daytime concentration in the classroom showed somewhat higher correlations.

An similarly designed study[14] among 37 non-smoking, non-occupationally exposed, 50- to 70-year-old adults, living in Amsterdam showed a median Pearson's R between personal and ambient PM10 concentrations of 0.50. Excluding days that subjects were exposed to ETS increased the correlation to a median R of 0.71, comparable to the value of 0.73 found in this study for non-ETS exposed children. In the Total Human Environmental Exposure Study (THEES), the correlation within subjects was calculated, using nine to 14 personal measurements from 13 non-smoking adults[9]. Individual personal-outdoor correlations ranged from 0.14 to 0.90 with a median value of 0.53. Using activity data improved the personal PM10 estimates for all individuals, to a median R of 0.93 (range 0.58 to 0.999). Exposure to ETS was one of the variables that contributed to this improvement, together with house-cleaning activities, cooking and use of unvented kerosene space heaters. Correlations after accounting for exposure to ETS alone were not described. Wallace[10] presented additional analyses of the PTEAM pilot study, which included repeated measurements of personal, indoor and outdoor PM10 in nine households

(two persons in each household). Cross-sectionally, personal PM10 exposures were uncorrelated with outdoor concentrations but for the 10 subjects with six to eight measurements, individual correlations ranged from -0.17 to 0.79, with a median value of 0.26.

Assuming that personal PM10 measurements are the most accurate estimate of the subject's true exposure, the correlation between personal and outdoor concentrations can be used to estimate the bias in the relationship between exposure and disease caused by using outdoor concentrations as a measure of exposure instead of personal exposures. If the measurement error in the exposure is non-differential and is the only source of error in the measure of the association between exposure and health effect, the relationship between the 'true' regression coefficient ($ß_t$) and the observed regression coefficient ($ß_o$) can be estimated as $ß_t = ß_o / R^2$.[15] With the median R of 0.6 found in our study, this implies that use of outdoor concentrations would result in a three-fold underestimation of the relationship between exposure and disease. This reasoning, however, strongly depends on the assumption that personal PM10 concentrations are the best measure of the relevant exposure. If not PM10 mass but fine particles or a specific component in PM10 is the causal agent responsible for the observed health effects, personal PM10 mass may not necessarily be the best exposure estimate.

The median slope between personal and outdoor concentrations of about 0.6 in our study was comparable to the slopes found for non-smoking adults in Amsterdam[14] and in the THEES and PTEAM study[10]. Slopes were similar for children with smoking and non-smoking parents.

It has been argued that the low (cross-sectional) correlation between personal and outdoor exposure to particles makes associations between day-to-day variations in outdoor air pollution and health effects implausible. The significant correlation between outdoor and personal exposure within subjects, over time, found in this study documents, however, that short term increases in outdoor air pollution are reflected in increased personal exposures. This finding provides support for using fixed site measurements as a measure of exposure to PM10 in time series studies linking day-to-day variations in outdoor concentrations to day-to-day variation in health endpoints.

Difference between personal and ambient concentrations

Personal exposures were on average 67 $\mu g/m^3$ higher than corresponding outdoor concentrations. The major part of this difference could be attributed to exposure to

ETS, high PM10 concentrations in the classrooms and (indoor) physical activity.

Children exposed to parental smoking had personal exposures that were about 40 $\mu g/m^3$ higher than children with non-smoking parents. This value is within the range of a 25 to 45 $\mu g/m^3$ increase in PM2.5 concentrations in homes with smokers that was recently suggested by Wallace[10].

PM10 concentrations in classrooms were significantly higher than the corresponding outdoor concentrations. PM10 measurements conducted in 11 other primary schools in the Netherlands have confirmed this finding[16]. (Indoor) physical activity was a third important source of increased personal exposures. Both findings are probably a result of resuspension of particles caused by the activity of the children. Thatcher and Layton[17] studied the effect of resuspension by measuring different particle size ranges before and after several resuspension activities. Five and 30 minutes of normal activity by four people and two minutes of continuous walking and sitting by one person resulted in a two to four-fold increase of particles in the 5-10 μm size range. In the PTEAM study[18], an estimated 'dirt level' in homes was significantly associated with 24-hour-averaged personal and indoor PM10 concentrations. Dirt and dust levels were estimated on a seven-point scale (0 to 3 by halves) by two 'calibrated' technicians. A 12-24 $\mu g/m^3$ increase in PM10 concentrations per unit increase in the index was predicted.

Excess personal exposures compared to indoor or outdoor concentrations have been found in most personal exposure studies[10]. Resuspension of coarse particles by personal activities and proximity to particle-generating sources have been suggested as causes of this so-called "personal cloud"[10]. This study shows that for children both particle-generating sources (smoking) and resuspension are important factors, causing significant differences between personal and outdoor PM10 concentrations.

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5. Personal exposure to fine particles in children correlates closely with ambient fine particles*

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Abstract

To investigate the validity of ambient fine particle (FP) concentrations as a measure of exposure in epidemiological time-series studies, the association between personal and ambient concentrations, within subjects, over time was established. Repeated measurements of personal and ambient FP were conducted in a group of 13 children, living in Wageningen, the Netherlands. For each child separately, personal exposures were related to ambient concentrations in a regression analysis. The median Pearson's correlation coefficient (R) was 0.86. Personal FP concentrations were also highly correlated with ambient PM10 concentrations (median R=0.75). Personal FP concentrations were on average 11 $\mu g/m^3$ higher than ambient concentrations. After excluding measurements of children exposed to Environmental Tobacco Smoke (ETS) the difference was only 5 $\mu g/m^3$. The high correlations between personal FP and both ambient FP and PM10, found in this group of children, provide support for the use of ambient PM concentrations as a measure of exposure to FP in epidemiological time series studies.

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Introduction

Recent epidemiological studies have documented associations between the day-to-day variation of particulate matter (PM) in ambient air and the day-to-day variation of mortality, hospital admissions and respiratory symptoms[1-3]. In these studies, exposure assessment was based on measurements conducted on fixed sites in ambient air. Measurements of personal exposure are considered a more accurate estimate of the subject's true exposure[4]. It has been suggested that PM concentrations measured at fixed sites in ambient air correlate poorly with personal exposures[5]. If the variation in outdoor concentrations is not tightly linked to the variation in personal exposures, use of outdoor concentrations as a surrogate for personal exposures would tend to misclassify personal exposures and exposure-response relationships could be attenuated[6]. In most personal exposure studies, however, the correlation between personal and ambient concentrations was calculated cross-sectionally. This correlation is influenced by the variation in personal exposures between subjects. As epidemiological time series studies relate day-to-day variations in ambient concentrations to day-to-day variations in health endpoints, it is more relevant to consider the correlation between personal and ambient concentrations within subjects, over time.

Recent epidemiological studies have often used PM10 measurements as the basis of exposure estimation[1]. PM10 consists of two size fractions: fine and coarse particles. Fine particles (FP) are usually defined as particles smaller than 2.5 μm (PM2.5), whereas coarse particles as larger than 2.5 μm. Both size fractions have different inhalation properties and chemical compositions[1,7]. Recently, it has been suggested that PM2.5 more than coarse particles (PM10 minus PM2.5) are specifically responsible for the observed associations between particulate matter air pollution and health effects[8]. At present, no information is available about the within-subject correlation between personal and ambient fine particles concentrations.

To investigate the validity of ambient PM concentrations as a measure of exposure to FP in time series studies, we conducted a personal exposure study in which repeated measurements of personal and ambient FP were conducted to allow calculation of the correlation between ambient and personal measurements, within subjects, over time.

Methods

Study design

Repeated measurements of personal and ambient fine particles (FP) were conducted in a group of 10- to 12-year-old children. Children were recruited through a primary school in Wageningen, a non-industrial town of about 35,000 inhabitants. Children were asked to participate after a presentation of the study in their classroom, which included a demonstration of the sampling equipment. The children received a written description and an informed consent form for their parents. We received parental permission for 15 out of 24 children (63%). One child dropped out after the first measurement and one child was excluded because she moved during the period of measurements. All remaining 13 children successfully completed the study.

Measurements took place from 29 March to 15 June, 1995. Eight measurements per child were planned. 24 hour averaged measurements of personal FP were conducted on weekdays only, spaced approximately one week apart. Samplers were distributed and collected at school, except for the first measurement when samplers were distributed to the children's homes to provide individual instruction to the children in the presence of a parent. Indoor measurements were conducted in the classroom. Outdoor measurements were conducted at a fixed site (see below).

Information on general characteristics such as parental smoking and housing conditions was collected by questionnaire. In addition, parents were asked to fill out a more detailed questionnaire about each day of measurements, including questions on exposure to Environmental Tobacco Smoke (ETS), time spent in several micro-environments and physical activity. After each day of measurements, these questionnaires were collected from the children's homes and checked for completeness and accuracy of the answers.

Sampling methods

Personal measurements were conducted with Casella respirable dust cyclones (Casella Ltd. London, UK), at a flow rate of 4 l/min. At this flow rate the cyclone measures particles with a 50% cut-off of approximately 3 μm. Air was sampled through 25 mm diameter 3 μm pore size Gelman Teflon filters, using a flow-controlled battery operated pump (Gilian, model Gil-air 5). Details about the sampling method and quality control issues are given elsewhere[9].

Outdoor measurements were conducted with the personal sampler at a fixed monitoring site. The site was located in a meadow on the outskirts of the town about 500 meters away from the nearest road, and at about 1.5 km distance from the school. No local PM sources such as unpaved roads, construction work or industries were present. Measurements were conducted at 1.5 meter height. In addition, hourly PM10 data were obtained from the Wageningen site of the National Air Quality Monitoring Network. At this site, PM10 measurements are conducted using a β-radiation monitor[10]. The site was located at 0.7 km distance from the FP monitoring site and at 1 km distance from the school.

Measurements of PM2.5 in the classroom were conducted with a Harvard PM2.5 Impactor (ADE Inc, Harrison, Maine, USA)[11]. A flow-controlled pump (ADE Inc, model SP-280E) and Anderson 37 mm 2 μm pore size Teflon filters were used. Measurements were conducted at 1.5 m height, away from the door and the blackboard to avoid disturbances by air currents and dust sources. Two averaging times were used: 24 hour measurements at the same time as the personal measurements (±15.00-15.00) and 7 hour measurements at the time the children were at school (±8.30-15.30). Co-located operation of the personal cyclone with the Harvard Impactor in the classroom for 12 days showed highly correlated concentrations (R=0.96) and no significant differences in the concentrations obtained with the two methods[9].

For the personal and classroom measurements, flows were measured at the beginning and end of each sampling period with calibrated rotameters and elapsed time indicators were used to calculate the sampled volume. For the ambient measurements sampled volumes were determined with a calibrated dry gas meter.

Personal and outdoor filters were weighed using a Mettler MT 5 micro-balance; classroom filters were weighed on an analytical balance with reading to the nearest 10 μg. All filters were weighed twice after equilibrating at 20 °C and 44% relative humidity for 24 hours, using a desiccator. Mean field blank weight changes were subtracted from all sample weights. The limit of detection, defined as 3 times the standard deviation in field blanks divided by the sampled volume, was 5.3 $\mu g/m^3$ for the personal and outdoor measurements. For the classroom measurements the detection limit was 3.7 $\mu g/m^3$ for the 24 hour averaged measurements and 12.7 $\mu g/m^3$ for the 7 hour average.

Data Analysis

The correlation between personal and ambient FP concentrations was assessed by means of individual regression analysis, with the equation:

$$FP_{personal,i\,t} = \alpha_i + \beta_i \times FP_{ambient,\,t} \qquad \text{Where } i = \text{child } i \text{ and } t = \text{day } t$$

The distribution of the individual regression results was investigated. Because of the limited number of observations per child used to calculate the individual coefficients, precision of the individual estimates is probably low. Most value should therefore be put on the population mean and median instead of the individual values. Medians are presented because not all coefficients were normally distributed. All children were non-smokers. No selection on parental smoking was made, however. Furthermore, even children with non-smoking parents could be exposed to ETS elsewhere or at home in the case of a visitor who smoked. The influence of exposure to ETS on the relationship between personal and ambient FP was investigated by excluding children with smoking parents and occasional days that children with non-smoking parents were exposed to ETS. The same analyses were conducted using the ambient PM10 concentrations measured by the National Air Quality Monitoring Network, instead of the ambient FP concentrations.

For comparison purposes, we calculated what the correlation would have been, if it had been calculated cross-sectionally. In this analysis, we randomly selected one measurement per subject and next calculated the cross-sectional correlation between personal and ambient FP concentrations. This procedure was repeated 1,000 times and the median of those 1,000 correlation coefficients was calculated to get a more reliable estimate of the cross-sectional correlation.

The questionnaire data were used to examine to what extent differences between personal and ambient concentrations could be explained by variables, such as exposure to ETS. The difference between personal and ambient concentrations was used as the dependent variable in a regression analysis. The SAS (Statistical Analysis System) procedure "PROC MIXED" was used to take correlations between repeated measurements of the same subjects into account. A random intercept model was used.

Results

13 children, six boys and seven girls, successfully completed the study. The average age was 10.8 years (range 10 to 12). Four children had a parent who smoked.

A total number of 77 personal measurements was obtained. 19 samples of the first two days of measurements were lost caused by damage of the filters. On the remaining 6 days of measurements, 92.1% of the measurements succeeded[9]. The individual mean personal and ambient FP concentrations are presented in table 1. Personal exposures were higher than ambient concentrations, especially for children exposed to parental smoking. After excluding four days on which children with non-smoking parents were occasionally exposed to ETS, the mean personal FP concentrations was 22.8 $\mu g/m^3$ (sd 2.6; range 18.7 to 26.0); about 5 $\mu g/m^3$ higher than mean of corresponding ambient concentrations (17.6 $\mu g/m^3$). Exposure to ETS was the only variable that was significantly related with the difference between personal and ambient concentrations. All other variables studied (physical activity, gender, time spent in a vehicle, cooking, cleaning activities) did not have a significant effect. The estimated effect of exposure to ETS was 23.8 $\mu g/m^3$ (se 4.6). The intercept of the model was 2.8 and not significantly different from zero.

Table 1. Distribution of individual averages of personal and ambient FP

	n	(#*)	Personal ($\mu g/m^3$)			Ambient ($\mu g/m^3$)		
			Mean	Sd	Range	Mean	Sd	Range
Non smoking parents	9	(55)	24.4	4.9	18.7 to 33.2	17.1	2.6	15.0 to 21.8
Smoking parent(s)	4	(22)	37.0	17.4	20.7 to 60.0	17.1	3.7	13.6 to 20.9
All children	13	(77)	28.3	11.3	18.7 to 60.0	17.1	2.8	13.6 to 21.8

* *Number of measurements*

Results of the FP measurements in the classroom are presented in table 2. FP concentrations in the classroom during school hours were about 5 $\mu g/m^3$ higher than the 24 hour averaged classroom concentrations. FP concentrations in

classrooms did not significantly differ from ambient concentrations. Table 3 shows the correlation matrix of classroom FP, ambient FP and ambient PM10 concentrations. Classroom concentrations were highly correlated with ambient concentrations.

Table 2. FP concentrations in the classroom ($\mu g/m^3$, n = 12)

	Mean	Sd	Range
7 hours*	19.9	5.8	14.1 to 35.2
24 hours, HI*	14.6	6.1	7.1 to 27.6
24 hours, PC†	15.4	6.2	8.7 to 29.0
Ambient†	16.8	11.8	6.2 to 45.2

* *Harvard Impactor*

† *Personal Cyclone*

Table 3. Pearson's correlation between classroom FP, ambient FP and ambient PM10 concentrations (n = 12)

	24 hours, HI*	24 hours, PC†	Ambient FP†	Ambient PM10‡
7 hours*	0.84	0.87	0.88	0.68
24 hours, HI*		0.96	0.96	0.89
24 hours, PC†			0.93	0.82
Outdoor FP†				0.90

* *Harvard Impactor*

† *Personal cyclone*

‡ *Measured with a β-radiation monitor by the National Air Quality Monitoring Network*

Correlation between personal and ambient concentrations

Results of the individual regression analyses for the relationship between personal and ambient FP concentrations are presented in table 4. Median Pearson's R was 0.86 for all children and 0.92 for children with non-smoking parents, after excluding four days with occasional exposure to ETS. Figure 1 shows the relationship between personal and ambient FP for non-ETS exposed children.

Median Pearson's R for the correlation between personal FP and ambient PM10 was 0.75 (range -0.25 to 0.97) for all children and 0.84 (range 0.67 to 0.96) for non-ETS exposed children.

The median value of 1,000 cross-sectional Pearson's correlation coefficients between personal and ambient FP was 0.41 (range -0.28 to 0.93) for all children and 0.82 (range -0.21 to 0.98) for non-ETS exposed children.

Table 4. Distribution of individual regression results, model $FP_{personal} = FP_{ambient}$

	All children (n = 13)		Non-ETS exposed* (n = 9)	
	Median	Range	Median	Range
Intercept	11.3	2.74 to 62.8	10.7	4.01 to 18.6
Slope	0.70	-0.19 to 1.67	0.53	0.44 to 1.00
Pearson's R	0.86	-0.11 to 0.99	0.92	0.63 to 0.97

* *children with non-smoking parents, after excluding days with occasional exposure to ETS*

Table 5. Distribution of children's individual correlation coefficients between personal and ambient PM10 and FP

	All children			Non-ETS exposed*		
	n	Median	Range	n	Median	Range
Personal FP - Ambient FP	13	0.86	-0.11 to 0.99	9	0.92	0.63 to 0.97
Personal FP - Ambient PM10	13	0.75	-0.25 to 0.97	9	0.84	0.67 to 0.96
Personal PM10 - Ambient PM10†	45	0.63	0.02 to 0.98	25	0.73	0.07 to 0.99

* *children with non-smoking parents, after excluding days with occasional exposure to ETS*

† *Measured in a different period and population*[12]

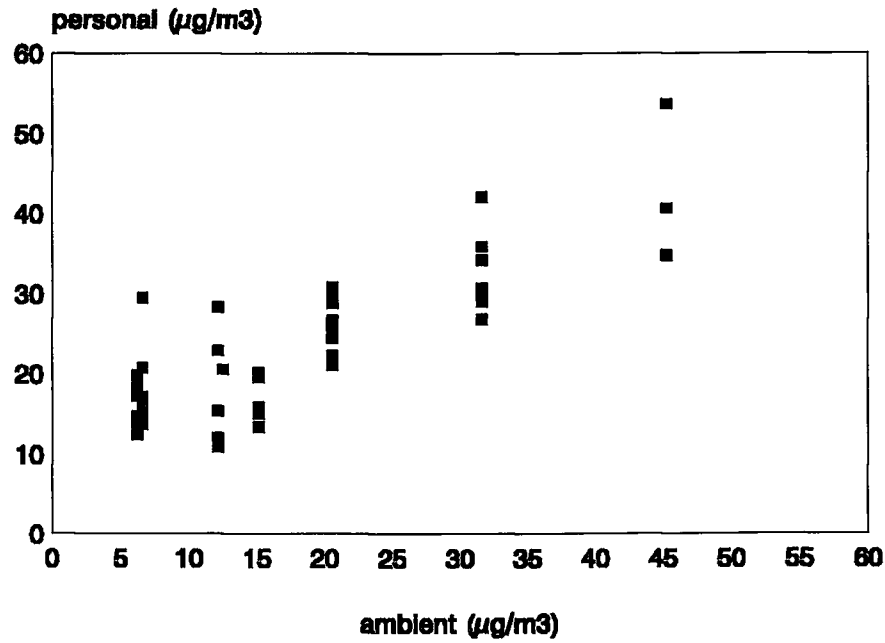

Figure 1. Relationship between personal and ambient FP for children with non-smoking parents, after excluding days with occasional exposure to ETS (n = 51)

Discussion

This study has shown that personal FP concentrations are highly correlated with ambient FP concentrations, within children, over time. The median of the individual correlation coefficients was 0.86 for all children and 0.92 for non-ETS exposed children. The estimated cross-sectional correlation coefficient for all children was considerably smaller (R=0.41).

Some other recent studies have shown reasonably high correlations between personal and ambient PM10 concentrations, within subjects, over time[12-14]. In an equally designed study among 45 10- to 12-year-old children we found a median Pearson's R between personal and ambient PM10 of 0.63 for children with non-smoking parents and 0.59 for children exposed to parental smoking. Excluding days that children with non-smoking parents were exposed to ETS resulted in a higher median R of 0.73[12]. In a similar study among 37 non-smoking, non-occupationally exposed, 50- to 70-year-old adults, we found a median Pearson's R between personal and ambient PM10 of 0.50. Excluding days that subjects were exposed to ETS increased the correlation to a median R of 0.71[13]. In both studies, correlations within subjects were higher than the estimated cross-sectional correlations. In the Total Human Environmental Exposure Study (THEES), Buckley *et al.*[14] also calculated the correlation between personal and ambient PM10 within subjects, using 9 to 14 personal PM10 measurements from 13 non-smoking adults. The median of the individual correlation coefficients was 0.53. Using time-activity data improved the personal PM10 estimates for all individuals, to a median R of 0.93. Exposure to ETS was one of the activity variables that contributed to the improvement of the individual correlations, together with house-cleaning activities, cooking and use of unvented kerosene space heaters.

These within-subject correlations for PM10 are lower than the correlation for FP found in this study. Little information is available about the within-subject correlation for FP. Wallace[15] presented additional analyses of the Particle Total Exposure Assessment Methodology (PTEAM) pilot study, which included repeated 12 hour averaged measurements of personal, indoor and ambient PM10 and PM2.5 in 9 households (2 persons in each household). Cross-sectionally, personal exposures were not correlated with ambient concentrations. The relationship between personal exposures and ambient concentrations improved when individual regression analysis was performed; for the 10 subjects with 6 to 8 individual measurements, the individual correlations ranged from -0.17 to 0.79 for PM10

and -0.17 to 0.76 for PM2.5, with a median value of 0.26 and 0.35 respectively. In these analyses, however, both daytime and night-time measurements were included. For the ambient measurements daytime and night-time concentrations were similar but daytime personal concentrations were higher than night-time personal concentrations[16,17]. The pooled analyses of daytime and night-time measurements may therefore have resulted in lower correlations than would be obtained if daytime and night-time concentrations could have been analyzed separately.

Although the cross-sectional correlations were lower than the median individual correlations, the cross-sectional correlations were also reasonably high, especially for the non-ETS exposed children (R=0.82). This seems inconsistent with the poor correlations between personal and ambient PM2.5/RSP found in other personal exposure studies[15]. All children attended the same class and 11 children lived in similar types of houses, in the same city district, within a radius of 1 km; only two children (one exposed to parental smoking) lived in another part of the city. Differences between these children were therefore probably smaller than differences between subjects in most other personal exposure studies, especially for the non-ETS exposed children. This is illustrated by the small variation (sd 2.6 $\mu g/m^3$) in the average personal exposures of non-ETS exposed children.

Personal FP concentrations were on average about 11 $\mu g/m^3$ higher than the corresponding ambient concentrations. For non-ETS exposed children the difference was only about 5 $\mu g/m^3$. FP concentrations in classrooms did not significantly differ from ambient concentrations. Recent studies on personal exposures to PM10 have found larger differences between personal and outdoor concentrations[12-15,19]. In the equally designed study on children's exposure to PM10[12] personal exposures were on average 67 $\mu g/m^3$ higher than the corresponding ambient concentrations (ambient mean was 39 $\mu g/m^3$). Exposure to ETS contributed about 20 $\mu g/m^3$ to this difference. Another 24 $\mu g/m^3$ could be explained by high PM10 concentrations in the classrooms: In the 3 schools where PM10 measurements were conducted, classroom concentration during school-hours were on average 123, 46 and 102 $\mu g/m^3$ higher than ambient concentrations. An important part of the remaining difference could be attributed to physical activity. The high classroom concentrations and influence of physical activity found for PM10 (and not for FP) are probably a results of re-suspension of coarse particles caused by the activity of the children. Thatcher and Layton[18] studied the effect of re-suspension by measuring different particle size ranges

before and after several re-suspension activities. 5 and 30 minutes of normal activity by four people, and 2 minutes of continuous walking and sitting by one person resulted in a two to fourfold increase of particles in the 5-10 μm size range, whereas particles in the 1-5 μm size range and sub-micron particles were hardly affected. In the PTEAM study[19], dirt and dust levels in homes were estimated on a seven-point scale (0 to 3 by halves) by two 'calibrated' technicians. An increase of 12-24 $\mu g/m^3$ in PM10 concentrations per unit increase in the index was predicted, whereas no effect on indoor PM2.5 concentrations was found. Elemental analysis of all personal and indoor PM10 filters, using XRF, showed that 14 or the 15 elements were uniformly elevated by values of 50-100% in the personal filters compared to the indoor filters. Only sulfur, which is mainly associated with sub-micron particles, was not elevated. The correlation between personal and ambient concentrations for sulfur was high ($R=0.88$), compared to a much lower correlation of 0.35 between personal and ambient mass concentrations[19]. These studies suggest that, apart from proximity to particles generating sources such as ETS, an important part of the so-called 'personal cloud', observed in other personal exposure studies[15], is caused by re-suspension of coarse particles.

In ambient air, PM10 and PM2.5 are generally highly correlated in areas with few sources of coarse particles. In this study, the correlation between our ambient measurements of FP and ambient PM10 measurements conducted by the National Air Quality Monitoring Network was 0.90. In the Harvard Six-City Study the correlation between PM2.5 and PM15 ranged from 0.80 to 0.97 for 5 of the 6 cities; only in Topeka, a city with relatively high concentrations of coarse particles, the correlation was lower ($R=0.45$)[20]. In the Netherlands, we found a correlation of 0.94 between daytime PM10 and PM2.5 concentrations measured simultaneously at the same site[21].

If personal FP concentrations are highly correlated with ambient FP concentrations, the high correlation between ambient PM10 and PM2.5 will result in a high correlation between ambient PM10 and personal FP also. Due to the influence of re-suspension of coarse particles on personal PM10 concentrations, the correlation between ambient PM10 concentrations and personal FP will probably even be higher than the correlation between ambient PM10 and personal PM10 concentrations. Table 5 summarizes the correlations found in this study and in our study on childhood exposure to PM10[12]. As expected, the correlations between personal FP and ambient PM10 were higher than the correlations

between personal PM10 and ambient PM10. Since both studies were conducted in different periods and with different children, however, the correlations are not directly comparable.

In this study, the correlation between personal and outdoor FP concentrations was established in a group of 13 primary school children, living in a non-industrial town. The question arises whether the results can be generalized to children living in cities and to (elderly) adults. Since ambient PM2.5, concentrations are generally spatially uniform across urban areas because of the importance of long range transport[22,23], PM2.5 exposures are probably just as well characterized by a single monitoring site in a city as in they are in a small town. This is supported by the study on childhood exposure to PM10, in which we did not find consistent differences in either the correlations or slopes between children in Wageningen and children in Amsterdam[12]. For adults, personal–outdoor relationships might be different from those of children because of differences in time activity patterns. For PM10, however, we found similar within subject correlations for older adults (aged 50 to 70) and for children[12,13]. Furthermore, elderly and/or retired adults probably spent more time in their homes. In our study of PM10 exposures of older adults as well as in the THEES study[24], indoor-outdoor correlations within homes were high. We therefore expect that personal FP exposures within older adults, over time, will also be highly correlated with ambient FP concentrations, as they are for children. However, actual personal monitoring studies of PM2.5 in adults are needed to establish this correlation.

In summary, this study has shown that personal FP concentrations are highly correlated with ambient PM concentrations, within children, over time. This finding provides support for the use of fixed site PM measurements as a measure of exposure to FP in epidemiological time series studies linking day-to-day variations in ambient concentrations to day-to-day variations in health endpoints.

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6. Mass concentration and elemental composition of PM10 in classrooms*

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Abstract

To investigate the sources of high PM10 concentrations in classrooms, observed in a previous study on childhood exposure to PM10, and to study the correlation between classroom and outdoor concentrations of PM10 mass and elements, the elemental composition of PM10 samples collected simultaneously in classrooms and outdoors was measured. Measurements of PM10 were conducted in two schools and outdoors in Amsterdam, the Netherlands. Averaging time was 24 hours for the outdoor measurements and both eight hours (school time) and 24 hours for the classroom measurements. X-ray fluorescense (XRF) analysis was used to measure the elemental composition of 55 samples from 11 days on which measurements were conducted simultaneously in both classrooms and outdoors. For most elements, classroom concentrations were considerably higher than outdoor concentrations, especially during school hours. The highest classroom/outdoor ratios were found for the soil related elements Si, Ca and Ti. The only elements that were not elevated were S, Br, Pb and Cl, which are dominated by non crustal sources. For S, Br and Pb, which are generally associated with submicron particles, also significant correlations between classroom and outdoor concentrations and between the two classrooms were found. The other elements generally showed low correlations. The results show that the high PM10 concentrations observed in our classrooms are probably due to resuspension of coarse particles and/or suspension of soil material. Due to these excess (re)suspended coarse particles, the correlation between classroom and outdoor concentrations is lower for elements associated with coarse particles than for elements associated with fine particles. Since the general composition of PM10 in classrooms differs from the composition of PM10 in ambient air, the high PM10 mass concentrations in classrooms can probably not be directly compared with ambient air quality guidelines.

* *Occupational and Environmental Medicine (provisionally accepted)*

Introduction

Several recent studies describe high personal exposures to PM10 compared with corresponding outdoor concentrations[1-4]. In a study on childhood exposure to PM10 in the Netherlands, personal exposures were on average 67 $\mu g/m^3$ (3 times) higher than the corresponding outdoor concentrations[3]. An important part of this difference could be explained by high PM10 concentrations in classrooms, which occurred especially during school time: in the three schools where PM10 measurements were conducted, 8 hour average classroom concentrations during school hours were two to five times higher than outdoor concentrations and about two times higher than 24-hour average classroom concentrations. PM10 measurements conducted in 11 other primary schools in the Netherlands also showed highly elevated classroom concentrations during school hours[5].

In the Particle Total Exposure Assessment Methodology (PTEAM) study[1,2], daytime personal PM10 concentrations (mean 150 $\mu g/m^3$) were about 60% higher than corresponding outdoor and indoor concentrations (mean 95 $\mu g/m^3$). Analysis of the elemental composition of the samples, using XRF, showed that nearly all elements were also elevated by 50-100% in the personal concentrations compared with the indoor concentrations, suggesting that the excess personal exposure has the same general composition as the indoor aerosol. Furthermore, during night time - when people were sleeping for about 2/3 of the time - personal PM10 concentrations (mean 77 $\mu g/m^3$) were more similar to the overnight indoor (mean 63 $\mu g/m^3$) and outdoor (mean 86 $\mu g/m^3$) concentrations. Re-suspension of household dust caused by human activity was therefore suggested as one of the causes of the elevated personal exposures. Thatcher and Layton[6] studied the of effect re-suspension by measuring different ranges of particle size before and after several re-suspension activities. Normal activity by four people or continuous walking and sitting by one person resulted in a two to fourfold increase of particles in the 5-10 μm size range. Re-suspension of particles, caused by the presence and activity of about 30 children in the relative small volume of a classroom, can therefore be considered a likely cause of the high PM10 concentrations observed in our classrooms. Furthermore, both studies suggest that re-suspension of particles occurs predominantly in the coarse fraction of PM10: in the PTEAM study, the only element that was not elevated in the personal samples was S, which is generally associated with submicron

particles; in the study by Thatcher and Layton, particles in the 1-5 μm size range and submicron particles were hardly affected by normal activity or continuous walking and sitting. Consistent with this, measurements of PM2.5 in a classroom did not show significant differences between classroom and outdoor concentrations[7].

To further investigate the elevated PM10 concentrations observed in classrooms, we measured the elemental composition of PM10 samples collected simultaneously in two classrooms and outdoors. Furthermore, the correlations between ambient and classroom concentrations of PM10 mass and elements were evaluated. Elemental composition of particles can be used to interpret what sources are important. In addition, specific elements can be associated specifically with fine or coarse particles.

Methods

Study design

In the framework of a study on childhood exposure to PM10[3], repeated measurements of PM10 were conducted in two primary schools in Amsterdam. In these classrooms, 24-hour average measurements as well as 8 hour average measurements during school time were conducted. Outdoor measurements were conducted using a 24 hour averaging time only. XRF analysis was performed on filters from all days that measurements were conducted in both schools simultaneously and an outdoor measurement was conducted with the same measurement method as the classroom measurements. This concerned 55 samples from 11 days, collected between 23 January and 7 March 1995.

Sampling sites

The study was conducted in Amsterdam, the capital of the Netherlands, which has about 720,000 inhabitants and is situated about 25 km East of the North Sea. In the inner city of Amsterdam, air pollution levels are influenced primarily by emissions from motorised traffic and long distance transport. The industrial area of Amsterdam is relatively small and is located in Amsterdam -West and -North.

Classroom measurements were conducted in two schools in the inner city of Amsterdam. School 1 was built in 1954 and school 2 in 1926. In school 2,

the ground floor and attic had been reconstructed in 1985. Furthermore, the stairwell of school 2 had been re-painted just before the start of the study. Distance between the schools was 1 km. School 1 was situated about 150 m South of a road with a traffic intensity of 17,000 vehicles per day and school 2 about 75 m East of a road with a traffic intensity of 14,000 vehicles per day. Earlier work that we conducted near a motorway carrying about 130,000 vehicles per day showed that at distances of about 100 meters or more, PM10 levels are not significantly different from background levels measured at greater distances[5]. Therefore, no significant influence of local traffic on classroom concentrations was expected. In each school, measurements were conducted in the classroom of children who participated in the study on childhood exposure to PM10, i.e. the highest grade (age 10 to 12). Both classrooms had uncarpeted floors and were situated on the first floor. Measurements were conducted away from the door and the blackboard to avoid disturbances by air currents and dust sources.

Outdoor measurements were conducted in a park in the city centre, about 150 m away from the nearest road and away from local particle sources such as unpaved roads, construction work or industrial sources. The outdoor monitoring site was located at 3 km distance from school 1 and at 2 km distance from school 2. PM10 measurements conducted by the National Air Quality Monitoring Network at 4 urban background and 4 street sites throughout the Netherlands, showed highly correlated PM10 concentrations (R 0.81-0.99) and no substantial differences in the yearly averages of the various sites[8]. PM10 concentrations measured at the outdoor monitoring site were therefore considered to be representative of PM10 concentrations throughout the city.

Sampling methods

PM10 measurements were conducted with a Harvard Impactor (ADE, Harrison, Maine, USA)[9] operating at 10 l/min, using Anderson 37 mm 2 μm pore size Teflon filters. Measurements were conducted at 1.5 m height. For the classroom measurements, two averaging times were used: 24-hour measurements at the same time as the outdoor measurements (15.00-15.00) and eight hour measurements when the children were at school (8.00-16.00). Flows were measured at the beginning and end of each sampling period with calibrated rotameters. Elapsed time indicators were used to calculate the sampled volumes.

Filters were weighed on an analytical balance with 10 μg reading precision, after equilibrating at 20 ° C and 44% relative humidity for 24 hours in a desiccator. All classroom filters were weighed in duplicate. At both the outdoor site and in the classrooms, field blanks were prepared by assembling filters in an extra impactor, which was placed without a pump at the sites during the measurements. Mean field blank weight changes were subtracted from all sample weights. Detection limits, defined as three times the standard deviation in field blanks divided by the sampled volume, were 8.0 $\mu g/m^3$ for the outdoor measurements, 3.7 $\mu g/m^3$ for the 24-hour average classroom measurements and 11.1 $\mu g/m^3$ for the 8 hour average classroom measurements. All measurements were above the detection limit.

Analysis of the elemental composition

The elemental composition of the samples was analysed by energy-dispersive XRF (x-ray fluorescence) at the US EPA facility in Research Triangle Park (North Carolina, USA). For each element the uncertainty per sample was calculated based on several factors including the concentration of the element in the sample and the propagated uncertainty calculated for sampling and analysis parameters, such as the calibration uncertainty and the system stability. The uncertainty limit was calculated as three times the uncertainty. Uncertainty limits thus changed from sample to sample for each element. Only elements with concentrations higher than the uncertainty limit on at least 50% of both outdoor and indoor filters were included in the data analysis.

In addition to uncertainty limits, detection limits, defined as three times the standard deviation of field blanks divided by the nominal sample volume, were calculated. Three field blanks were analysed. Mean field blank values were subtracted from all sample values.

Results

Fourteen elements were measurable on at least 50% of both the outdoor and indoor filters. Mean field blank concentrations, detection limits and percentage above detection and uncertainty limits of those 14 elements are presented in table 1. Elements are grouped to their main ambient sources: S, Pb, Br (anthropogenic), Cl (marine), Cr, Zn, Cu, Mn, K, Fe (both crustal and anthropogenic) and Si, Ca, Ti, Sr (crustal)[10-13]. Values that were above the uncertainty limit were always also higher than the detection limit. For most elements, correcting for the mean field blank values did not result in a substantial change in the concentrations. For Br, however, the correction resulted in a more than 60% increase in the 8 hour average concentrations and an approximate 20% increase in the 24-hour average concentrations.

Table 1. Detection limits (DL), % above DL and % above Uncertainty limits (UL) for elements measurable on at least 50% of outdoor and classroom filters

	Mean field blank (ng/m^3)		DL (ng/m^3)		% > DL	% > UL
	School hours*	24 hour**	School hours*	24 hour**		
S	3.8	1.3	36.3	12.1	100	100
Pb	–2.4	–0.8	3.6	1.2	100	100
Br	–4.6	–1.5	3.7	1.2	98	87
Cl	–28.1	–9.4	15.8	5.3	100	100
Cr	–0.4	–0.1	3.0	1.0	96	89
Zn	2.4	0.8	4.1	1.4	100	100
Cu	–0.5	–0.2	5.7	1.9	100	100
Mn	–3.0	–1.0	5.0	1.7	100	98
K	42.3	14.1	102.9	34.3	100	100
Fe	9.3	3.1	20.6	6.9	100	100
Si	–92.6	–30.9	26.0	8.7	100	98
Ca	7.9	2.6	16.0	5.3	100	100
Ti	1.4	0.5	8.1	2.7	100	89
Sr	1.2	0.4	2.9	1.0	100	98

* *based on a sampled volume of 4.8 m^3*

** *based on a sampled volume of 14.4 m^3*

Mass and elemental concentrations are presented in table 2. Ratios between classroom and outdoor concentrations are shown in figure 1. Medians are presented because concentrations and ratios were not normally or log-normally distributed for all elements.

Table 2. Median mass and elemental concentrations of classroom and outdoor PM10 ($\mu g/m^3$ for mass, ng/m^3 for elements; n = 11)

	School 1			School 2			Outdoor
	School hours	24 hour	Non-school hours*	School hours	24 hour	Non-school hours*	24 hour
Mass	164.2	73.3	34.7	77.8	44.2	28.5	28.5
S	1,173	756	482	878	769	662	862
Pb	43	31	18	71	61	49	26
Br	14	7	6	13	8	6	11
Cl	1,759	1,571	1,537	2,893	1,553	1,288	1,499
Cr	19	8	2	17	9	6	2
Zn	165	77	46	142	90	61	24
Cu	34	22	17	19	13	10	6
Mn	66	27	14	16	9	6	8
K	1,439	648	338	536	319	189	140
Fe	2,135	934	500	614	442	341	209
Si	10,264	4,224	1,853	3,992	1,978	1,060	194
Ca	5,919	2,540	1,075	3,322	1,919	1,222	232
Ti	524	271	152	235	153	114	10
Sr	29	12	5	15	7	4	2

* *estimated classroom concentration during non-school hours;*

estimated $= (C_{24\ hour} \times t_{24\ hour} - C_{school\ hours} \times t_{school\ hours})/(t_{24\ hour} - t_{school\ hours})$

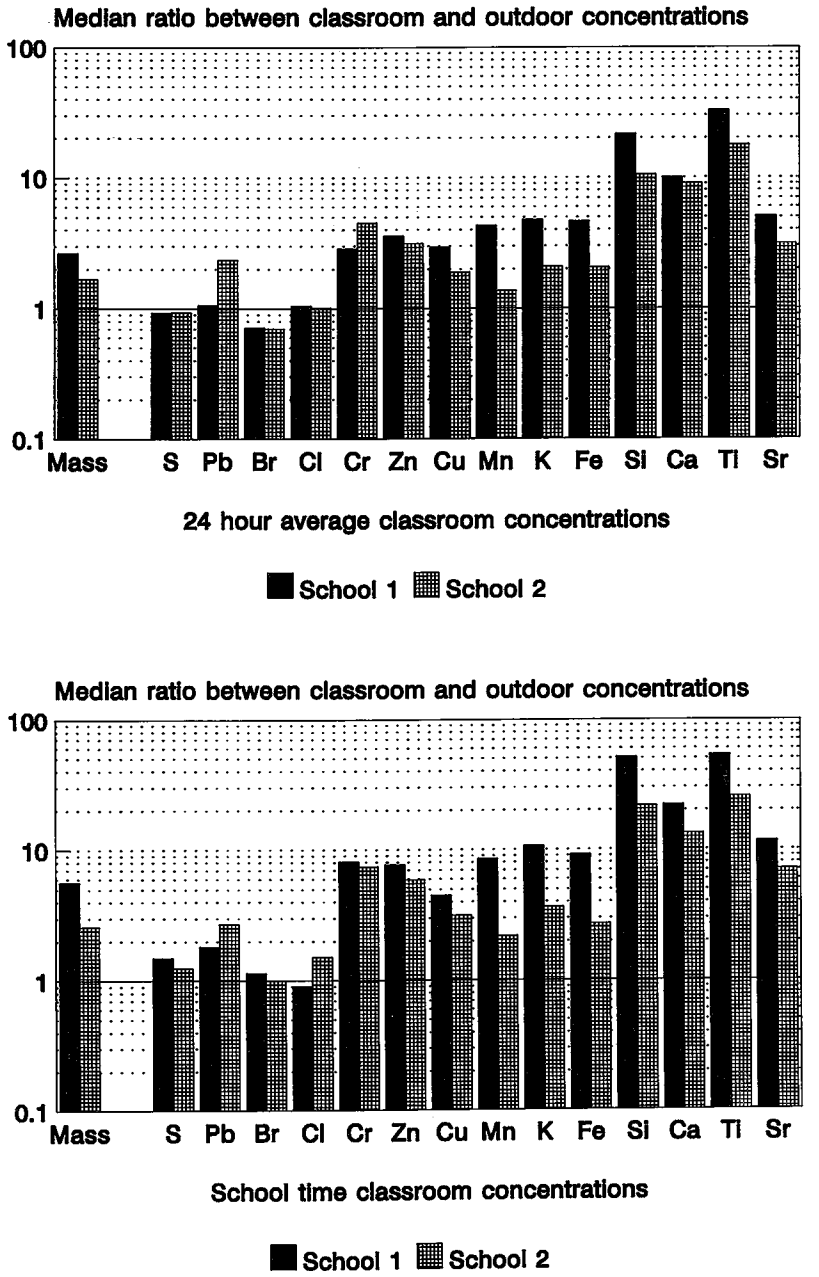

Figure 1. Median ratio between classroom and outdoor concentrations for 24-hour average (upper) and school time (lower) classroom concentrations

For most elements, classroom concentrations were considerably higher than outdoor concentrations. This was especially the case during school time. Hourly PM10 data of the same time period, obtained from the Amsterdam site of the National Air Quality Monitoring Network, did not show significant differences between schooltime and 24-hour average concentrations (mean 29.2 $\mu g/m^3$ and 29.6 $\mu g/m^3$, respectively). The differences between school time classroom measurements and outdoor concentrations can therefore not be explained by diurnal variation in ambient concentrations. The highest classroom/outdoor ratios were found for the soil related elements Si, Ca and Ti, with median classroom/outdoor ratios up to 50 for schooltime classroom Si and Ti concentrations in school 1. Elements that were classified to have both crustal and anthropogenic sources (Cr, Zn, Cu, Mn, K, Fe) showed schooltime classroom concentrations that were 2 to 11 times higher than outdoor concentrations. Only for S, Br and Cl, 24-hour average classroom concentrations were lower than or comparable to outdoor concentrations. For Pb, 24-hour average classroom concentrations were similar to outdoor concentrations in school 1, whereas much higher classroom concentrations were found in school 2. Mass concentrations in school 1 were higher than in school 2. This was also the case for most elements, especially for the soil-related elements.

Spearman correlations between classroom and outdoor concentrations, and between the two classrooms are presented in table 3. Figure 2 displays the relations between 24-hour average classroom and outdoor concentrations. Because outdoor measurements were only conducted using a 24-hour averaging time, the comparison between classroom and outdoor concentrations should focus on the 24-hour average classroom concentrations. Classroom mass concentrations were only moderately correlated with outdoor concentrations. A significant correlation was only found between outdoor and 24-hour average classroom concentrations in school 1. Mass concentrations in school 1 were not correlated with mass concentrations in school 2. For several crustal elements, 24-hour average concentrations in school 1 were also significantly correlated with outdoor concentrations, but other correlation coefficients were generally low and not significant. For S and Br, however, all concentrations that were measured with the same averaging time (i.e. between outdoor and 24-hour average classroom concentrations, and between the two classrooms) were significantly correlated. For Pb, significant correlations were found for school 1 but not for school 2.

Table 3. Spearman correlation between classroom concentrations and 24-hour average outdoor concentrations (n = 11)

	24-hour average classroom concentrations			8-hour average classroom concentrations during school hours		
	Outdoor – School 1	Outdoor – School 2	School 1 – School 2	Outdoor – School 1	Outdoor – School 2	School 1 - School 2
Mass	0.63*	0.43	–0.02	0.36	0.32	–0.05
S	0.84**	0.95**	0.76**	0.58†	0.54†	0.64*
Pb	0.95**	0.52	0.55†	0.66*	0.19	0.30
Br	0.75**	0.85**	0.74**	0.23	0.53†	0.67*
Cl	0.46	0.95**	0.39	0.27	0.64*	0.64*
Cr	0.45	–0.31	–0.15	0.39	0.00	–0.29
Zn	0.81**	0.34	0.12	0.52	0.32	0.43
Cu	–0.02	0.58†	0.35	0.40	0.05	0.04
Mn	0.63*	0.61*	0.45	0.56†	0.42	0.70*
K	0.72*	0.14	0.12	0.65*	–0.01	0.08
Fe	0.63*	0.51	0.38	0.50	0.07	0.50
Si	0.63*	0.05	–0.08	0.72*	–0.19	0.07
Ca	0.63*	–0.43	–0.34	0.40	–0.41	0.14
Ti	0.49	0.29	–0.18	0.51	–0.27	–0.09
Sr	0.26	0.32	–0.06	0.29	0.15	–0.22

† $p < 0.10$

* $p < 0.05$

** $p < 0.01$

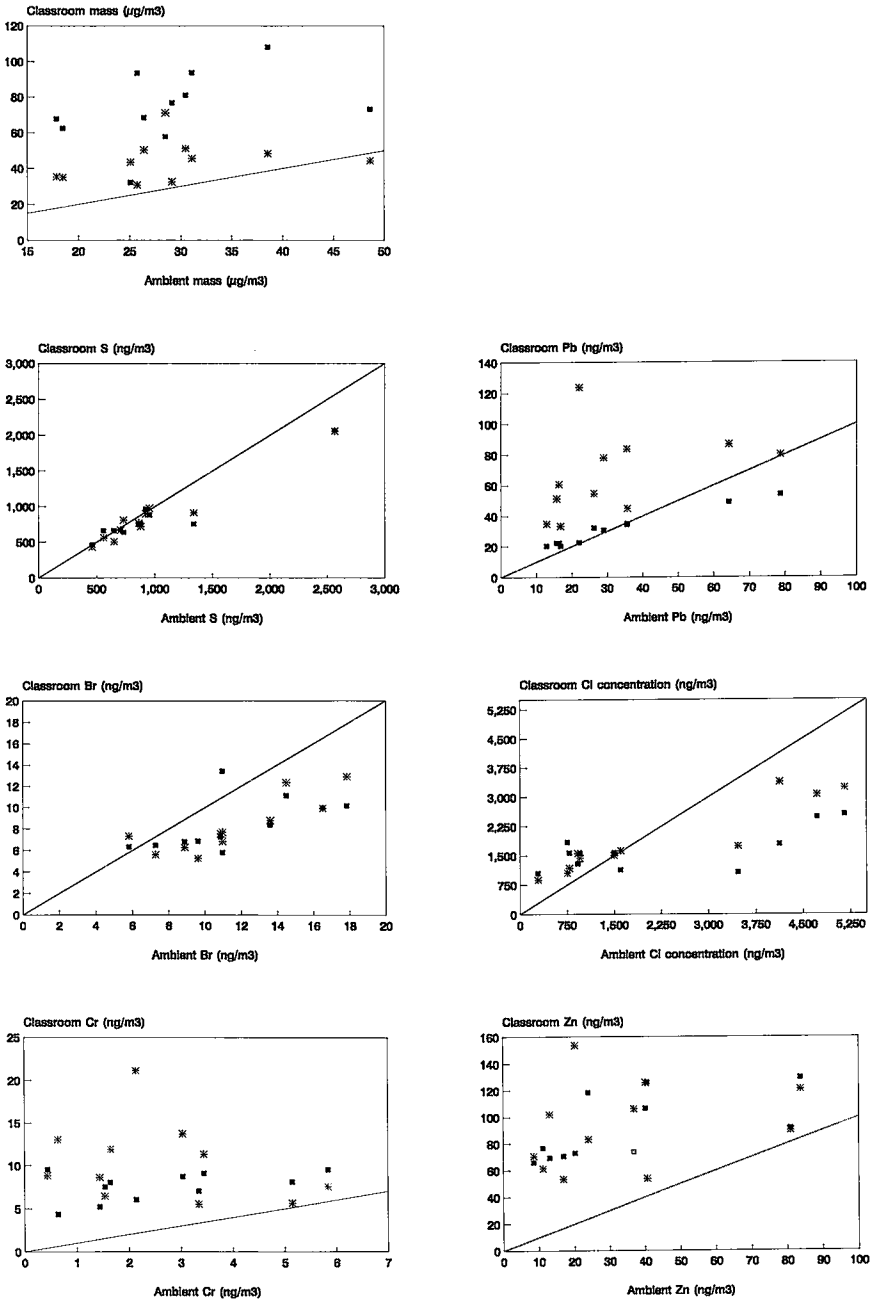

Figure 2. Relation between 24-hour average outdoor and classroom concentrations (■ = school 1, ✳= school 2; line shows 1:1 line)

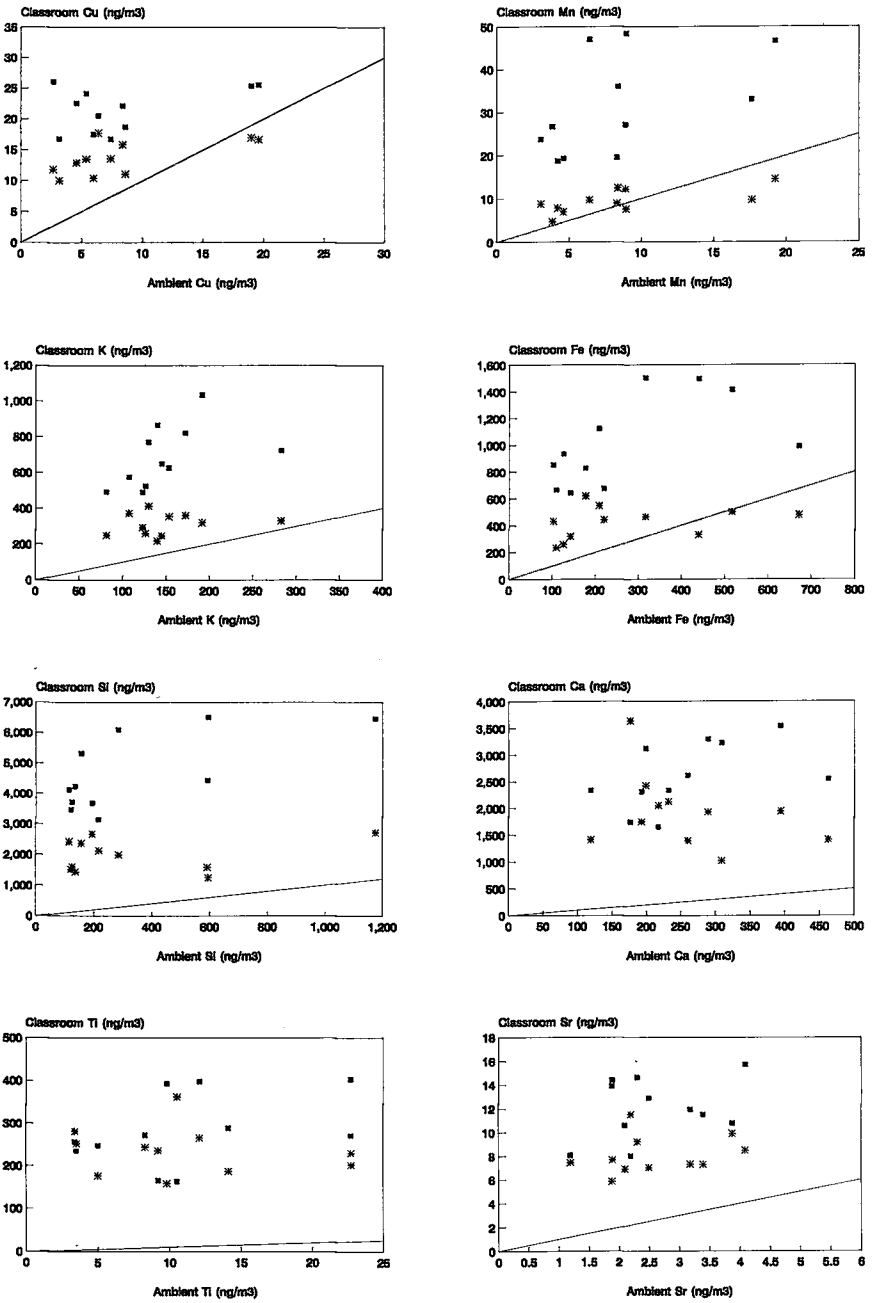

Figure 2 (continued). Relation between 24-hour average outdoor and classroom concentrations (■ = school 1, ✳= school 2; line shows 1:1 line)

Discussion

This study has shown that mass concentrations and most elemental concentrations of PM10 in classrooms were considerably higher than outdoor concentrations, especially during school hours. The highest classroom/outdoor ratios were found for the soil related elements Si, Ca and Ti. The only elements that showed 24-hour average classroom concentrations lower or equal to the corresponding outdoor concentrations were S, Br, Pb (school 1 only), which are dominated by combustion sources, and Cl, which is dominated by marine aerosol. Given the re-painting that had been conducted in school 2 just before the start of the study, the high Pb concentrations in school 2 were possibly caused by removal of old lead-based paint.

Limited information is available about particulate matter (PM) concentrations in classrooms. Roorda-Knape *et al.*[5] also found highly elevated PM10 concentrations in 11 schools in the Netherlands. The average schooltime PM10 concentrations in the classrooms ranged from 51 to 145 $\mu g/m^3$, compared with an outdoor PM10 concentration of about 26 $\mu g/m^3$. Thompson *et al.*[15] measured indoor and outdoor 24-hour average TSP concentrations in 16 buildings in California, including 6 schools. Indoor concentrations were lower than outdoor concentrations in all buildings with air filtration. In the three schools without air filtration, indoor concentrations exceeded outdoor concentrations (I/O ratio of 1.04, 1.15 and 3.82 respectively). PM2.5 measurements conducted in one classroom in the Netherlands[7] did not show significant differences between classroom and outdoor concentrations. Mean classroom concentrations were 20 $\mu g/m^3$ and 15 $\mu g/m^3$ for schooltime and 24-hour average measurements respectively, compared with an average outdoor concentration of 17 $\mu g/m^3$.

Several studies in homes and offices have generally found lower indoor/outdoor ratios for mass and crustal elements than the ones found in our study[16-18,1-2]. Two studies in closed, unoccupied rooms[16] or sites selected to have a minimum of activity likely to lead to re-suspension[17] found average indoor/outdoor ratios well below one (range 0.1 to 0.5) for Pb, Br, Zn, Fe and Ca, measured in TSP using XRF. In a study in 25 homes and 5 office buildings in Finland[18], daytime measurements were conducted of both fine (<1.5 μm) and coarse ($>$ 1.5 μm) particles, and the elemental composition (Si, S, Cl, K, Ca, Fe, Cu, Zn) was measured using Particle Induced X-ray Emission (PIXE). For fine

particles, the highest indoor/outdoor ratios (range 1.2 to 4.8) were found for elements originating from indoor sources (Cl and K). In the homes, elevated indoor/outdoor ratios (range 1.5 to 2.8) were also found for Ca and Fe, but these were explained to be due to a few exceptionally high values. For coarse particles, elevated indoor/outdoor ratios (range 1.4 to 4.8) were found in the homes for mass, Cu, Zn and Cl, but not for Si, Ca and Fe (range 0.4 to 0.8). In the PTEAM study[1,2], mass and elemental concentrations of PM10 in the 178 homes were generally lower or similar to outdoor concentrations, both during daytime and during nighttime. During nighttime, this was also the case for personal exposure measurements. For daytime personal samples, however, the pattern of results was more similar to our study: mass concentrations (mean 150 $\mu g/m^3$) were considerably higher than outdoor and indoor concentrations (mean 95 $\mu g/m^3$), and, except for S, all elements measured were also elevated by 50-100% in the personal concentrations compared with the indoor concentrations. Nevertheless, these elevations are small compared with the ones found in our classrooms, especially for Si, Ca and Ti.

In the classrooms, no particle emitting sources such as smoking, woodburning and gas stoves were present. The high classroom concentrations are also not likely to be caused by local traffic: the highest concentrations were found in school 1, which was situated at greater distance and much less frequently downwind of the nearest busy road than school 2. Furthermore, the highest classroom concentrations were found during school hours, when the classrooms were occupied by approximately 30 children. The most probable cause of the elevated classroom concentrations, therefore, is re-suspension of settled dust and/or suspension of soil material brought in by the children's shoes. Use of chalk for writing on the blackboard could also be dust source. Chalk consists mainly of Calciumcarbonate and will therefore predominantly affect Ca concentrations. Since the classroom/outdoor ratios for Si and Ti were higher than for Ca, chalk dust was probably not the most important source of the high PM10 concentrations. Thatcher and Layton[6] studied the effect of resuspension by measuring different particle size ranges before and after several resuspension activities. 5 and 30 minutes of normal activity by four people, and 2 minutes of continuous walking and sitting by one person resulted in a two to fourfold increase of particles in the 5-10 μm size range. Particles in the 1-5 μm size range and sub-micron particles, however, were hardly affected. In the PTEAM study[2], dirt and dust levels in homes were estimated on a seven-point scale (0 to

3 by halves) by two 'calibrated' technicians. An increase of 12-24 $\mu g/m^3$ in PM10 concentrations per unit increase in the index was predicted, whereas no effect on indoor $PM_{2.5}$ concentrations was found. Raunemaa *et al.*[18] found that coarse particle (>1.5 μm) concentrations depended approximately linearly on the time spent indoors, whereas no such relationship was found for fine particles (<1.5 μm). These studies suggest that human activity contributes primarily to re-suspension of coarse particles. In our study, the highest classroom/outdoor ratios were found for soil related elements and the lowest ratios were found for elements dominated by combustion sources. Since soil related elements are generally associated with coarse particles and elements from combustion sources are generally associated with submicron particles[19,20], this is consistent with the general finding that re-suspension activities mainly affect coarse particle concentrations.

The largest classroom/outdoor ratios were found for Si, Ca and Ti. These elements showed large differences in their relative contribution to classroom and outdoor mass concentrations. In total Si and Ti contributed about 6% to the schooltime classroom mass concentrations, compared with less than 1% to the outdoor mass concentrations. Since these elements can be considered as the most specific soil elements, this suggests that the elevated classroom concentrations are not only caused by re-suspension of settled dust, but also by suspension of soil material.

The estimated concentrations during non-school hours were also elevated compared with the outdoor concentrations, especially for Si, Ca and Ti. After schooltime, classroom concentrations remain higher than outdoor concentrations until the excess (re)suspended particles are deposited. Thather and Layton[6] stated that in case of much higher indoor than outdoor concentrations, the deposition loss rate can be calculated using the following equation:

$$\lambda_d = t^{-1} \times \ln(C_i/C) - \lambda_v \qquad (1)$$

where λ_d is the deposition rate (h^{-1}), C_i is the initial concentration ($\mu g/m^3$), C is the final concentration ($\mu g/m^3$), t is the time between C_i and C (h), and λ_v is the air exchange rate (h^{-1}). Consequently, the time it takes till classroom concentrations are back to outdoor levels can be estimated as:

$$t = \ln(C_i/C_{outdoors}) \times (\lambda_d + \lambda_v)^{-1} \qquad (2)$$

Thatcher and Layton[6] calculated a deposition rate of 0.46 h^{-1} for particles in the 1-5 μm range and 1.36 h^{-1} for particles in the 5-10 μm range. In the PTEAM

study the estimated deposition rate for PM10 mass was 0.65 h^{-1}. Deposition rates for Si, Ca and Ti in PM10 were within the same range (0.60-0.63)[2]. No information was available about the air exchange rates in the classrooms we studied. During non-school hours, the schools were closed and unoccupied. Furthermore, both schools had the policy to close all windows after schooltime. Air exchange rates during non-school hours were therefore probably low. The sum of the deposition and air exchange rate ($\lambda_d + \lambda_v$) can therefore be expected to have a value in the range of 1 to 2 h^{-1}. Using the median concentrations during schooltime as the initial concentrations, it can be estimated that for example for Si and Ti it took 2 to 4 hours in school 1 and 1.5 to 3 hours in school 2 until classroom concentrations were decreased to outdoor levels. Directly after schooltime, it is likely that concentrations were considerably higher than the 8 hour average concentrations due to (re)suspension caused by the children leaving the classroom and daily sweeping of the classroom floor immediately after schooltime, so the time it takes until outdoor concentrations are reached is probably longer. These calculations show that it is plausible that concentrations during non-school hours were still higher than outdoor concentrations. Because of the many uncertainties and assumptions, however, a quantitative statement about the extent to which the elevated concentrations during non-schools hours were a consequence of high concentrations directly after schooltime can not be made.

The highest correlations between classroom and outdoor concentrations and between classrooms were found for S, Br and Pb (school 1). These elements have in common that they all had 24-hour average classroom concentrations lower or similar to the corresponding outdoor concentrations and originate primarily from anthopogenic sources, which are generally associated with submicron particles. For Cl, classroom/outdoor ratios were also low, but concentrations were less well correlated than for S, Br and Pb. Cl differs from these elements in that it has marine aerosol as its major source, which is generally associated with coarse particles. Elements that were elevated in the classroom samples compared with the outdoor samples, generally showed low correlations. In the PTEAM study[2], S was the only element that was not elevated in the personal samples compared with indoor samples. S was also the only element for which personal and indoor concentrations were highly correlated with outdoor concentrations (R^2 0.8-0.9). All other elements showed lower correlations between personal and outdoor concentrations (R^2 0.1-0.4).

The correlation coefficient (R) between daytime personal and ambient mass concentrations was 0.35. Several other studies on personal, indoor and outdoor sulfate, have also found high correlations, with correlation coefficients between personal and outdoor concentrations or between indoor and outdoor concentrations ranging from 0.81 to 0.99[21-24]. In one of these studies[21], mass concentrations of respirable suspended particles (RSP) were also measured. The correlation between personal and outdoor RSP concentrations (R 0.69) was lower than between personal and outdoor sulfate (R 0.81). In a study on childhood exposure to fine particles[7], classroom concentrations were similar to and highly correlated ($R>0.90$) with ambient concentrations. Evidently, when personal or indoor concentrations are significantly influenced by (re)suspension of predominantly coarse particles, these excess coarse particles reduce the correlation between personal or indoor concentrations and outdoor coarse particle concentrations, whereas correlations for fine particles are less influenced.

Recently, it has been suggested that fine particles (PM2.5) more than coarse particles (PM10 minus PM2.5) are specifically responsible for the observed associations between particulate matter air pollution and mortality[25]. In addition, it has been hypothesised that the (metal)composition of the particles affects the associated health response[26]. In this case, indoor PM10 measurements in environments with a lot of activity would not provide a good measure of the relevant exposure. Furthermore, since the general composition of PM10 in classrooms differs from the composition of PM10 in ambient air, the high PM10 mass concentrations in classrooms can not be directly compared with ambient air quality guidelines.

Conclusion

This study has shown that the high PM10 mass concentrations, observed in a previous study on childhood exposure to PM10, are probably due to resuspension of coarse particles and/or suspension of soil material, caused by the activity of the children. Due to these excess (re)suspended coarse particles, the correlation between classroom and outdoor concentrations is lower for coarse particles (and associated elements) than for elements that are generally associated with fine particles. Since the general composition of PM10 in classrooms differs from the composition of PM10 in ambient air, the high PM10

mass concentrations in classrooms can probably not be directly compared with ambient air quality guidelines.

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7. General discussion

Main findings

Personal PM10 exposures of both adults (aged 50 to 70 years) and children (aged 10 to 12 years) were reasonably well correlated over time with ambient PM10 concentrations. Personal FP exposures were highly correlated with ambient FP concentrations (table 1).

Table 1. Average levels of personal and outdoor concentrations, and the correlation (R) between personal and outdoor concentrations

Population	Size fraction	n	Mean Personal* ($\mu g/m^3$)	Mean Ambient* ($\mu g/m^3$)	Median individual R	Cross-sectional R†
All subjects						
Adults	PM10	37	62	42	0.50	0.34
Children	PM10	45	105	39	0.63	0.28
Children	FP	13	28	17	0.86	0.41
Non-ETS exposed						
Adults	PM10	23	51	41	0.71	0.50
Children	PM10	25	89	40	0.73	0.49
Children	FP	9	23	18	0.92	0.84

* Mean *of individual averages*

† *Estimated cross-sectional R, by randomly selecting 1 measurement per subject*

After excluding days with exposure to ETS, correlation coefficients increased. In all cases, the medians of the individual correlation coefficients were higher than the estimated cross sectional correlation coefficients. Personal FP concentrations were also highly correlated with ambient PM10 concentrations: the median R was 0.75 for all subjects and 0.84 for non-ETS exposed subjects (chapter 5).

Personal exposures exceeded outdoor concentrations. An important part of these differences could be attributed to exposure to ETS. For adults, an additional part of the difference between personal and outdoor concentrations could be attributed to living along a busy road and time spent in traffic (chapter

3). After excluding measurements of subjects exposed to ETS, differences between personal and outdoor concentrations were relatively small for PM10 in adults (10 $\mu g/m^3$) and for FP in children (5 $\mu g/m^3$). Personal PM10 concentrations among non-ETS exposed children, however, were still more than two times higher than ambient PM10 concentrations. An important part of this remaining difference could be attributed to high PM10 concentrations in classrooms, which occurred especially during school time (chapter 4). Results of the analysis of the elemental composition of the PM10 samples in 2 of the 3 classrooms suggest that these high classroom concentrations were due to re-suspension of coarse particles and/or suspension of soil material, caused by the presence and activity of the children (chapter 6). Indoor physical activity was found to be a third factor that was significantly associated with elevated PM10 exposure in children. This influence of physical activity on personal exposures is probably also caused by (re)suspension of particles. After taking the influence of exposure to ETS, physical activity and the high PM10 concentrations in classrooms into account, the major part of the difference between personal and outdoor PM10 concentrations in children was explained.

Comparison with other studies

Correlation between personal and outdoor concentrations

Few other studies have investigated the correlation between personal and outdoor PM concentrations within subjects. In the Total Human Environmental Exposure Study (THEES), Buckley *et al.*[1] calculated the correlation within subjects, using 9 to 14 repeated personal PM10 measurements from 13 non-smoking adults in Phillipsburg, NJ. The median of the individual correlation coefficients was 0.53 (range 0.14 to 0.90). Using activity data improved all correlations to a median R of 0.93. Exposure to ETS was one of the variables that contributed to this improvement, together with house cleaning activities, cooking and use of unvented kerosene space heaters. Correlations after accounting for exposure to ETS alone were not described. Wallace[2] presented both the cross-sectional and within-subjects correlations using data from 14 subjects in the THEES study. The cross-sectional correlation between personal and outdoor concentrations was 0.52, whereas the median of the individual correlations was 0.68. Wallace[2] also presented additional analyses of the Particle Total Exposure Assessment Methodology (PTEAM) pilot study, which

included repeated 12 hour averaged measurements of personal and outdoor PM10 and PM2.5. Cross-sectionally, personal exposures were not correlated with ambient concentrations. For the 10 subjects with 6 to 8 individual measurements, however, the median of the individual correlations was 0.26 for PM10 and 0.35 for PM2.5. As in our study, these studies report higher individual correlations compared to cross-sectional correlations. Although the findings from the PTEAM pilot study also suggest higher correlations for PM2.5 than for PM10, the correlations are considerably lower than the correlations found in our study and in the THEES study. In the analyses of the PTEAM pilot data, however, both daytime and night-time measurements were included. For the ambient measurements daytime and night-time concentrations were similar, but daytime personal concentrations were higher than night-time concentrations[3,4]. The pooled analyses of daytime and night-time measurements may therefore have resulted in lower correlations than would be obtained if daytime and night-time concentrations could have been analysed separately or averaged to 24 hour data.

Difference between personal and outdoor concentrations

Higher personal exposures to PM compared with indoor and outdoor concentrations have been found in most personal exposure studies[2]. Exposure to ETS is considered to be one of the most important sources of excess personal or indoor particle concentrations[2]. The estimated contributions of exposure to ETS found in our study are within the range of values found in other studies, as recently reviewed by Wallace[2]. Re-suspension of particles by personal activity has been suggested as a second important cause of elevated personal exposures[2]. Several studies found that human activity can cause elevated indoor particle concentrations, and they suggest that human activity contributes mostly to re-suspension of coarse particles[5-7]. For example, Thatcher and Layton[5] found that 5 and 30 minutes of normal activity by four people, and 2 minutes of continuous walking and sitting by one person resulted in a two to fourfold increase of particles in the 5-10 μm size range, whereas particles in the 1-5 μm size range and submicron particles were hardly affected. In our study, the highest classroom/outdoor ratios of elemental concentrations of PM10 samples were found for soil related elements (Si, Ti, Ca) and the lowest ratios were found for elements dominated by combustion sources (S, Br, Pb). Since soil related elements are generally associated with coarse mode particles and elements from combustion sources are generally associated with submicron particles[8,9], this is

consistent with the general finding that re-suspension activities mainly affect coarse particle concentrations. The small differences between personal and outdoor FP concentrations, and between classroom and outdoor FP concentrations further support this theory.

Potential biases and limitations

In this section, first several aspects that could have biased the observed relationships between personal and outdoor concentrations will be discussed. Second, some limitations with regard to the generalisability of the results will be considered.

Measurement methods and timing

For children (both PM10 and FP), personal and outdoor concentrations were directly comparable since measurements were conducted with the same measurement methods and all children in one school received and returned the equipment at approximately the same time, which coincided with the start and end of the outdoor measurements. For logistic reasons, however, this design was not possible for the adults: different samplers were used for personal and outdoor measurements, and personal samplers were distributed and collected throughout the day whereas outdoor sampling was conducted from 15.00 – 15.00. A field comparison, however, showed high correlations and no significant differences between outdoor concentrations obtained with the different measurement methods (chapter 2). With regard to the sampling times, the average overlap between the measuring periods of personal and outdoor measurements was large: 21 hours. The minimum overlap of 18 hours occurred when the personal sampler was distributed and collected at 9.00. Calculation of 24 hour averaged PM10 concentrations both from 15.00 to 15.00 and from 9.00 to 9.00, using hourly PM10 measurements conducted in Amsterdam by the National Air Quality Monitoring Network, showed highly correlated 24 hour averaged concentrations (R 0.96). Therefore, although the use of different sampling methods and not completely overlapping sampling times for personal and outdoor sampling in adults may have resulted in somewhat lower correlation coefficients, this bias is probably small.

Ambient monitoring site

Ambient concentrations were characterised using measurements conducted at a single monitoring site in the city or town where the subjects lived. Concentrations of PM10 and especially PM2.5 generally show little spatial variation across urban areas[7,10,11]. In the Netherlands, PM10 measurements conducted by the National Air Quality Monitoring Network at 4 urban background and 4 street sites throughout the country, showed highly correlated 24-hour averaged PM10 concentrations (R 0.81 – 0.99) and no substantial differences in the yearly averages at the various sites[12]. In the city of Arnhem, we found high correlations between daytime concentrations measured simultaneously at a street and background site for both PM2.5 and PM10 (R 0.97 and 0.92, respectively), despite a small but significant difference in the concentration level[13]. The day-to-day variation in ambient PM concentrations across a city or town can therefore be considered to be well characterised by measurements at a single outdoor monitoring site.

Response and compliance

Response rates were low, especially in Amsterdam; only 39 out of 195 adults (20%) who had already agreed to participate in a panel study on acute effects of air pollution on respiratory health[14], and 15 out of 56 children (27%) could be included in the personal exposure study. In Wageningen, response rates were higher (58%, children only). The main objective of the study was to evaluate the relation between personal and ambient PM concentrations, within subjects, over time. In this case, the low response can only have biased the results in the unlikely case that the relation between personal and ambient particles for the participants differs from the relation for subjects who were not included. Because conducting personal measurements is very labour intensive and repeated measurements for each subject were necessary, compliance was considered more important than response. We therefore did not attempt to optimise the response, but instead tried to minimise the drop-out during the course of the study. After the first measurement, therefore, participants were explicitly asked whether they were willing to carry the monitor another seven days. This resulted in the drop-out of a considerable number of adults (12 adults = 24%), but resulted in a high compliance for the remaining volunteers: 95% of the subjects who agreed to continue participating after the first measurement, both adults and children, successfully completed the study.

Personal exposure measurements are not only labour intensive, but also

intrusive for the study participants. Concern, therefore, exists that wearing a monitor can cause the participant to change his or her behaviour and consequently introduce bias[15]. The possibility that subjects would tend to stay at home or spend less time outdoors due to the wearing of the personal monitor was evaluated by comparing time activity on days of personal sampling with time activity on other weekdays (chapter 2). For children, no significant differences between sampling and non-sampling days were found. Adults, however, spent significantly less time outdoors and more time at home on days of personal sampling, compared to other weekdays. The absolute differences in the adults' time activity on days of personal sampling compared with other weekdays, however, were rather small: on average + 0.9 hours for time spent at home and -0.5 hours for time spent outdoors. For particle mass concentrations, it is unlikely that such a small shift in time activity will cause large differences in the exposure measured. Furthermore, when the average differences in time activity between sampling and non-sampling days were used to divide the subjects into two groups, no considerable differences in the regression results of the two groups were found (chapter 3). The change in behaviour therefore probably did not influence the relation between personal and outdoor/indoor PM10.

Indoor sources

In our study, the correlation between personal and ambient mass concentrations of PM10 and FP was investigated. Personal and indoor mass concentrations were found to be significantly influenced by indoor sources. In studies that relate ambient PM air pollution to health effects, however, not the total personal exposure to particles but personal exposure to ambient particles is most relevant[11]. In this case, associations between the day-to-day variations in health endpoints and the day-to-day variation in ambient PM can only be confounded by indoor particle generating activities in case indoor-generated PM concentrations are statistically dependent on outdoor PM concentrations[11]. It is not likely that subjects, for example, will smoke more or conduct more other particle generating activities on days with high PM air pollution than on days with low PM air pollution. It has been suggested, however, that if study subjects would close their windows on days with higher levels of pollution, exposure to indoor pollutants might increase and actually be responsible for the increase in adverse health outcomes and therefore confound the particle and health effect association[16]. In our study, since no smog warnings were issued during the study

period, it is unlikely that subjects have closed their windows in direct response to high PM air pollution. However, in the winter period, the highest ambient PM concentrations occur on days with low temperature and/or low wind speed[12], which could indirectly cause lower ventilation rates on days with high air pollution than on days with low air pollution. Hoek *et al.*[17] measured indoor RSP and NO_2 concentrations, before, during and after an air pollution episode in The Netherlands in the winter of 1984/1985. During the episode, indoor concentrations were higher than on non-episode days. The increase was observed in all three categories of homes (0 smokers, 1 smoker, 2 smokers). The authors therefore concluded that the increases in indoor air concentrations observed during the episode were largely due to penetration of air pollution from outdoors, and not due to decreased ventilation, leading to increased concentrations of pollutants generated indoors. In a study of personal PM10 exposure among Japanese elderly subjects[18,2] the influence of indoor sources was probably low: all subjects were non-smokers, not living with smokers and measurements which were affected by a visitor's smoking or by burning of incense and/or mosquito coils were excluded from the analysis. Furthermore, the influence of re-suspension was probably low due to the relative inactive time activity patterns of elderly subjects in general, and the relative cleanliness of the Japanese homes caused by the use of traditional 'tatami' mat flooring and the habit of removing shoes before entering. This study differs from most other personal PM10 studies not only in that personal exposures (mean 37 $\mu g/m^3$) were lower than outdoor concentrations (mean 56 $\mu g/m^3$), but also in that personal and outdoor PM10 concentrations were highly correlated (cross-sectional R 0.83).

The relation between outdoor concentrations and personal exposure to ambient particles can be further evaluated by considering components that are predominantly of outdoor origin, such as sulphur or sulphate. In our study, S was one of the few elements for which classroom concentrations were similar to and highly correlated with ambient concentrations (chapter 6). In the PTEAM study[7], S was the only element that was not elevated in the personal PM10 samples compared with indoor samples, and S was also the only element for which personal and indoor concentrations were highly correlated with outdoor concentrations (R^2 0.8 to 0.9). All other elements showed lower squared correlations between personal and outdoor concentrations (R^2 0.1 to 0.4). The correlation coefficient (R) between daytime personal and ambient PM10 mass concentrations was 0.35. Several other studies on personal, indoor and outdoor

sulphate have also found high cross-sectional correlations, with correlation coefficients between personal and outdoor concentrations or between indoor and outdoor concentrations ranging from 0.81 to 0.99[19-22]. In one of these studies[19], mass concentrations of personal respirable suspended particles (RSP) were also measured. The correlation between personal and outdoor RSP mass concentrations (R 0.69) was lower than between personal and outdoor sulphate (R 0.81). The correlations found for PM10 and FP mass concentrations therefore probably underestimate the correlation between outdoor concentrations and personal exposure to ambient particles.

Weekdays versus weekends

Personal exposure measurements were only conducted on weekdays. In weekends, personal-outdoor relationships could be different because of differences in time activity patterns. For example, children do not spend any time at school in the weekend and are therefore likely to spend more time outdoors or in other locations. In our study, however, the average time spent outdoors by children in weekends was only about 0.5 hour longer than on weekdays. For adults, the average time spent outdoors was similar in weekends compared to non-sampling weekdays. In a study on time activity patterns conducted in Ede, The Netherlands[23,24], the average time spent outdoors of 5 to 12 year old children and working adults (aged 21-65 years) in the winter season was only about 1 hour longer in weekends compared with weekdays. For non-working 21-65 year old adults and subjects aged over 65+, the average time spent outdoors did not differ between weekdays and weekends. Although the differences between time spent outdoors in weekends compared with weekdays appear to be small, it is not known to what extent the allocation of time over various types of indoor environments differs between weekends and weekdays, and, more importantly, between different weekends. It is not unlikely that time activity patterns of especially children and working adults will be more variable in weekends. The correlation between personal and outdoor concentrations may therefore be lower in weekends than on weekdays. In some types of epidemiological time series studies, measurements, of for example pulmonary function[25], are also only conducted on weekdays, so the correlations found in our study at least hold for this kind of studies. For studies that included both weekdays and weekends, for example on daily peak flow or respiratory symptoms, the applicability in this respect may be somewhat limited.

Locations

PM10 measurements were conducted in the city of Amsterdam as well as in a small town (Wageningen, children only). For FP, measurements were only conducted in Wageningen. Since ambient PM2.5 and PM10 concentrations are generally spatially uniform across urban areas without major local point sources[7,10,11], PM2.5 and PM10 exposures are probably just as well characterised in a city as they are in a town. This is supported by the results of the PM10 measurements in children, where we did not find consistent differences in either the correlations or slopes between children in Wageningen and children in Amsterdam (chapter 4). The results of the study can therefore be generalised to many other cities and towns.

Population

Personal PM10 measurements were conducted in 50- to 70-year-old adults and primary school children (aged 10 to 12 years). FP measurements were added to the study later and were only conducted in a small group of children. The question arises whether the results can be generalised to other populations. Personal-outdoor relationships might be different for different population groups because of differences in time activity patterns. For PM10, however, we found similar within subject correlations for non-ETS exposed older adults compared to non-ETS exposed children. When compared with children, many other population groups probably spend more time in their homes. In our study of PM10 exposures of older adults (chapter 3) as well as in the THEES study[26], correlations between indoor and outdoor concentrations were high for most individual homes. We therefore expect that personal FP exposures within subjects who spend the major part of their time at home will also be highly correlated over time with ambient concentrations, as they are for children. For working adults, however, personal–outdoor correlations may be different because of potential occupational exposures. For mortality studies, the correlation between personal and outdoor concentrations for subjects who are most likely to die because of air pollution is relevant. If this would primarily involve critically ill hospitalised subjects, the relevant personal-outdoor relationships for these studies could be different because of the relatively clean environment of a hospital, especially when air-conditioning or air filtration systems are used. Schwartz[27], however, found that the major part of excess deaths on days with high air pollution occurred outside a hospital. The same pattern was observed in the London episode of 1952[27]. The results of this study

can therefore probably be generalised to many populations studied in time series studies on acute health effects of air pollution. For some population groups, however, specific personal monitoring studies are needed to establish the correlation between personal and outdoor concentrations.

Season

Measurements were conducted in autumn and winter for adults, and in winter and spring for children. No measurements were conducted in the summer period. In the study on time activity patters conducted in Ede[23,24], time spent outdoors was higher in summer than in winter for all population groups (difference 0.9 to 2 hours on weekdays and 1.4 to 3.3 hours in weekends). In addition, air exchange rates will be higher in summer because windows and doors will be opened more often. The fraction of outdoor particles found indoors will therefore be higher in summer than in winter. As a result of the longer time spent outdoors and the higher infiltration of particles, the correlation between personal and outdoor concentrations will probably be better in summer than in the seasons that the study was conducted.

Implications

It has been argued that the low (cross-sectional) correlation between personal and outdoor exposure to particles makes associations between day-to-day variations in outdoor air pollution and health effects implausible. The significant correlation between outdoor and personal exposure within subjects, over time, that we found documents, however, that short term increases in outdoor air pollution are reflected in increased personal exposures. This finding provides support for using fixed site measurements as a measure of exposure to PM in time series studies linking day-to-day variations in outdoor concentrations to day-to-day variation in health endpoints.

Recently, it has been suggested that fine particles are more likely to be responsible for the observed associations between PM air pollution and health effects than the coarse part of PM10[16,11]. It has been hypothesised that the associations found between day-to-day variation in PM10 and health are in fact the result of an underlying relation with FP, because the variation in ambient PM10 serves as an index for the variation in ambient FP[7,11]. Schwartz *et al.*[28] compared effect estimates of the association between daily mortality and PM10,

PM2.5 as well as coarse particles (CP = PM10 minus PM2.5). PM10 and PM2.5 were both significantly associated with increased mortality, while no association was found with CP, suggesting that fine particles and not coarse particles were specifically responsible for the observed associations between particulate matter air pollution and mortality in this study. Apart from potential differences in the pathogenic properties, the fraction of outdoor air particles present in indoor air differs between fine and coarse particles[11]. This fraction depends on the penetration factor, the air exchange rate and the particle deposition rate[2]. Recent studies have shown that the penetration factor is close to one for all particles less than 10 μm in aerodynamic diameter[5,7]. The deposition rate, however, increases with increasing particle size[5]. Personal exposures to ambient particles will be therefore higher for fine particles than for coarse particles[11]. For example, Wallace[2] estimated that the fraction of outdoor particles found indoors under equilibrium conditions, at an air exchange rate of 0.76 h^{-1}, will be 66% for PM2.5 and 43% for the coarse part of PM10 (PM10 minus PM2.5).

In our study, for children both PM10 and FP were measured. Results for the two fractions differ on two major points:

1. For PM10, personal and classroom concentrations considerably exceeded outdoor concentrations, whereas for FP these differences were much smaller.
2. For PM10, the correlation between personal and outdoor concentrations, within children, over time, was lower than for FP.

Since the personal FP and PM10 measurements were not conducted simultaneously on the same child, but in different groups of children and different time periods, these comparisons are indirect. Results of the XRF analysis of the classroom PM10 samples, however, show a similar pattern of results: elements that are generally associated with fine mode particles (S, Br, Pb) had classroom concentrations that were similar to/lower than and highly correlated with outdoor concentrations, whereas elements that are generally associated with coarse mode particles (Si, Ca, Ti) had extremely high classroom concentrations and generally showed lower correlations between classroom and outdoor concentrations. Other studies have also found higher correlations between personal and outdoor concentrations for sulphur or sulphate than for PM10 or RSP mass[7,19]. Evidently, when personal or indoor concentrations are significantly influenced by (re)suspension of predominantly coarse particles, these excess coarse particles reduce the correlation between personal or indoor concentrations and outdoor coarse particle concentrations, whereas correlations for fine particles are less influenced.

In ambient air, PM10 and PM2.5 are highly correlated in areas without major sources of coarse particles. In Wageningen, the correlation between our ambient measurements of FP and ambient PM10 measurements conducted by the National Air Quality Monitoring Network was 0.90 (chapter 5). In Arnhem, we found a correlation of 0.94 between daytime PM10 and PM2.5 concentrations measured simultaneously at the same site[13]. In the PTEAM study, the correlation between outdoor PM10 and PM2.5 was 0.89 and 0.97 for daytime and night-time measurements respectively[4]. In a study in metropolitan Philadelphia, the correlation between PM10 and PM2.5 was 0.95[10]. If personal FP concentrations are highly correlated with ambient FP concentrations, the high correlation between ambient PM10 and PM2.5 will result in a high correlation between ambient PM10 and personal FP also. In chapter 5 we showed that the correlation between ambient PM10 concentrations and personal FP was higher than the correlation between ambient PM10 and personal PM10 concentrations, found in another time period and another group of children. In case fine particles are indeed specifically responsible for the observed associations between PM air pollution and respiratory health effects, the high correlation between personal FP and outdoor PM10 concentrations found in our study provides stronger support for the use of outdoor PM10 as a measure of exposure in time series studies than the correlations found between personal PM10 and outdoor PM10.

If the correlation between the (most accurate estimate of the) 'true' exposure and a specific exposure estimate is known, this correlation can be used to estimate the bias in the relationship between exposure and disease caused by using the surrogate measure of exposure instead of the true exposure[29]. If the measurement error in the exposure is non-differential and is the only source of error in the measure of the association between exposure and health effect, the relationship between the 'true' regression coefficient ($ß_t$) and the observed regression coefficient ($ß_o$) can be estimated as $ß_t = ß_o / R^2$.[29] If personal FP concentrations are considered to be the most accurate estimate of the 'true' exposure, the correlations between personal FP exposures and outdoor concentrations of non-ETS exposed subjects found in this study would imply that use of outdoor FP concentrations (median R 0.9) would results in an about 20% underestimation, and use of outdoor PM10 concentrations (median R 0.84) in an about 40% underestimation of the relationship between exposure and disease. If personal PM10 concentrations are used as the 'golden standard', however, the median R of 0.7 would suggest a much larger underestimation of the true regression slopes (about 100%). These calculations show that the bias

in the relation between exposure and disease strongly depend on which exposure variable is considered to be the most accurate estimate of the 'true' exposure. If FP (or a specific component in PM air pollution more associated with fine than with coarse particles) is the causal agent responsible for the observed health effects, personal PM10 mass may not be the best exposure estimate.

The air quality standard for particles in the United States is based on the mass of the particles and is therefore not chemically specific[30]. Although there have been efforts to identify the role of biologically active chemical species, the processes that underlie the effects of PM air pollution on health are still poorly understood[30]. Nevertheless, it seems reasonable to suspect that chemical composition is a feature that determines the pathogenicity of the particles[16]. The associations between mass concentrations and health effects would then be the result of an underlying relationship with a specific component that is correlated with the ambient mass concentration. In indoor environments with indoor sources of particles, however, the composition of the particles can be considerably different from the composition of ambient particles. This was demonstrated by the XRF analysis of the classroom samples, which showed that for example Si and Ti contributed about 6% to the schooltime mass concentration of PM10, compared with less than 1% to the outdoor mass concentrations. In such cases, indoor PM10 mass concentrations can not be directly compared with ambient air quality guidelines.

Conclusions

This study has shown that personal PM10 concentrations of both children and older adults are reasonably well correlated with ambient concentrations, within subjects, over time. Personal FP concentrations are highly correlated with both ambient FP and PM10 concentrations. The lower correlations for PM10 are probably due to the larger impact of (re)suspension of coarse particles on PM10 than on FP. Correlations within subjects, over time, were higher than cross-sectional correlations. These findings provide support for the use of fixed site measurements as a measure of exposure to PM in epidemiological time series studies linking the day-to-day variation in PM to the day-to-day variation in health endpoints.

Personal PM exposures significantly exceeded outdoor concentrations,

especially for PM10 in children. These differences between personal and outdoor concentrations could be largely attributed to exposure to ETS and (re)suspension of coarse particles.

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Summary

This thesis describes a study of the relation between outdoor concentrations and personal exposure to particulate matter (PM) air pollution. Chapter 1 presents the background of the study. Recent epidemiological studies have documented associations between PM and several acute health effects. These studies are mostly time series studies, relating day-to-day variations in air pollution to day-to-day variations in health endpoints. In these studies, exposure assessment is generally based on measurements conducted on fixed sites in ambient air. It has been suggested that these fixed site measurements correlate poorly with personal exposures, which raises questions about the plausibility of the observed exposure-response relationships. In most personal exposure studies, however, the correlation between personal and outdoor concentrations was calculated cross-sectionally. For time series studies on acute health effects the correlation between personal and ambient concentrations within subjects, over time, is more relevant. Only limited information is available about the within-subject correlation between personal and outdoor PM concentrations. The first goal of this thesis therefore is to evaluate the relation between personal and ambient PM concentrations, within subjects, over time. The second goal is to evaluate potential differences between personal, indoor and ambient PM concentrations.

Repeated measurements of personal and outdoor PM concentrations were conducted in a series of studies with similar design. Personal PM10 measurements were conducted in 37 adults in Amsterdam and 45 primary school children from both Amsterdam and Wageningen. In addition, personal exposure to fine particles (FP) was measured in 13 children in Wageningen. Outdoor measurements were conducted at a fixed monitoring site in both Amsterdam and Wageningen. In addition, indoor measurements were conducted in the living rooms of the adults and the classrooms of the children. Seven to eight measurements per subject were planned. Averaging time was 24 hours for the personal, living room and outdoor measurements, and both 24 hour and 8 hour (school time) for the classroom measurements. For each subject separately, personal exposures were related to outdoor concentrations using linear regression analysis. The distribution of the individual correlation and regression coefficients was investigated. Furthermore, the extent to which differences

between personal and outdoor concentrations could be explained was studied using questionnaire data about factors that might influence personal exposures.

In chapter 2 the methods used to measure the personal exposure to PM10 and FP are described. Method performance was evaluated regarding compliance, sampling flow, weighing procedure, field blanks and co-located operation of the personal samplers with stationary methods. Furthermore, the possibility that subjects change their behaviour due to the wearing of personal sampling equipment was studied by comparing time activity on days of personal sampling with time activity on other weekdays. Compliance was high; 95% of the subjects who agreed to continue participating after the first measurement, successfully completed the study. Except for the first two days of FP sampling, over 90% of all personal measurements were successful. All pre and post sampling flow readings were within 10% of the required flow rate of 4 L/min. Precision of the gravimetric analyses was satisfactory. All concentrations were above the detection limits. Co-located operation of the personal sampler with stationary samplers showed highly correlated concentrations ($R>0.90$) and no considerable differences in concentrations obtained with the different methods. Adults spent significantly less time outdoors (0.5 hour) and more time at home (0.9 hour) on days of personal sampling compared to other weekdays. For children no significant differences in time activity were found.

Chapter 3 describes the relationships between personal, indoor and outdoor PM10 concentrations in 50- to 70-year-old adults. All adults were non-smokers, not living with smokers and with no occupational exposure to dust. Median Pearson's correlation coefficient (R) between personal and outdoor concentrations was 0.50. Excluding days with exposure to environmental tobacco smoke (ETS) improved the correlation to a median R of 0.71. The estimated cross-sectional correlations were lower; 0.34 and 0.50 respectively. Outdoor concentrations (mean 42 $\mu g/m^3$) exceeded indoor concentrations (mean 35 $\mu g/m^3$) but underestimated personal exposures (mean 62 $\mu g/m^3$). The major part of the difference between personal and outdoor concentrations could be attributed to exposure to ETS, living along a busy road and time spent in a vehicle.

Chapter 4 presents the relationship between personal, classroom and outdoor PM10 concentrations in 10- to 12 year-old children. Median R between personal and outdoor concentrations was 0.63 for children with non-smoking parents and 0.59 for children with smoking parents. The estimated cross-

sectional correlations were lower; 0.45 and 0.20, respectively. For children with non-smoking parents, excluding days with exposure to ETS improved the correlation to a median R of 0.73. The mean personal PM10 concentration was 105 $\mu g/m^3$; on average 67 $\mu g/m^3$ higher than the corresponding outdoor concentrations. The major part of this difference could be attributed to exposure to ETS, to high PM10 concentrations in the classrooms, and to indoor physical activity.

In chapter 5, the relationship between personal and outdoor FP in 10- to 12 year-old children is presented. Median R was 0.86. The estimated cross-sectional R was lower (0.41). Personal FP concentrations were also highly correlated with ambient PM10 concentrations (median R 0.75). Personal FP concentrations were on average 11 $\mu g/m^3$ higher than ambient concentrations. After excluding measurements of children exposed to ETS the difference was only 5 $\mu g/m^3$. FP concentrations in the classrooms were similar to, and highly correlated with, ambient concentrations.

In chapter 6, the causes of the high PM10 concentrations found in classrooms (chapter 4), and the correlation between classroom and outdoor concentrations of PM10 mass and elements are investigated. X-ray fluorescense (XRF) analysis was used to measure the elemental composition of samples of 11 days on which measurements were conducted simultaneously in two schools and outdoors in Amsterdam. For most elements, classroom concentrations were considerably higher than outdoor concentrations, especially during school hours. The highest classroom/outdoor ratios were found for the soil related elements Si, Ca and Ti. The only elements that were not elevated were S, Br, Pb and Cl, which are dominated by non crustal sources. For S, Br and Pb, which are generally associated with submicron particles, also significant correlations between classroom and outdoor concentrations and between the two classrooms were found. The other elements generally showed low correlations. The results show that the high PM10 concentrations observed in our classrooms are probably due to resuspension of coarse particles and/or suspension of soil material.

Chapter 7 presents the general discussion of the study. First, the main findings of the study are summarised and compared with findings of other studies. Next, some potential biases and limitations with regard to the generalisability of the study are discussed. Finally, the implications and conclusions of the study are given. The conclusion of this thesis is that personal

PM exposures are reasonably well (PM10) to highly (FP) correlated with ambient concentrations, within subjects, over time. The lower correlations for PM10 are probably due to the larger impact of (re)suspension of coarse particles on PM10 than on FP. Correlations within subjects were higher than cross-sectional correlations. These findings provide support for the use of fixed site measurements as a measure of exposure to PM in epidemiological time series studies linking the day-to-day variation in PM to the day-to-day variation in health endpoints. Personal PM exposures significantly exceeded outdoor concentrations, especially for PM10 in children. These differences between personal and outdoor concentrations could be largely attributed to exposure to ETS and (re)suspension of coarse particles.

Samenvatting

In dit proefschrift wordt een onderzoek naar de relatie tussen buitenluchtconcentraties en persoonlijke blootstelling aan stofvormige luchtverontreiniging beschreven. In hoofdstuk 1 wordt de achtergrond van het onderzoek gegeven. In recente epidemiologische onderzoeken zijn associaties gevonden tussen stofvormige luchtverontreiniging en een aantal acute gezondheidseffecten. Dit betreft veelal zogenaamde tijdreeksstudies, waarin de dag-tot-dag variatie in luchtverontreiniging wordt gerelateerd aan de dag-tot-dag variatie van gezondheidseindpunten. In deze studies wordt de blootstelling aan luchtverontreiniging gekarakteriseerd door middel van metingen op een vast meetpunt in de buitenlucht. Uit enkele onderzoeken blijkt echter dat deze op een vast meetpunt gemeten concentraties slecht correleren met de persoonlijke blootstelling, waardoor de plausibiliteit van de beschreven gezondheids-effect relaties in twijfel is getrokken. Deze interpretatie is echter gebaseerd op dwarsdoorsnedeonderzoek waarbij verschillende personen éénmalig zijn bemeten. Voor tijdreeksstudies naar de acute effecten van luchtverontreiniging is echter met name de correlatie tussen persoonlijke blootstelling en buitenluchtconcentraties, binnen personen, in de tijd, relevant. Er is weinig informatie beschikbaar over de binnenpersoonscorrelatie tussen persoonlijke blootstelling en buitenluchtconcentraties. De eerste doelstelling van dit proefschrift is daarom het evalueren van de relatie tussen persoonlijke blootstelling en buitenluchtconcentraties van stofvormige luchtverontreiniging, binnen personen, in de tijd. Het tweede doel is het evalueren van mogelijke niveauverschillen tussen persoonlijke, binnenlucht- en buitenluchtconcentraties.

Herhaalde metingen van zowel persoonlijke blootstelling als buitenluchtconcentraties zijn gedaan in een aantal studies met vergelijkbare opzet. Persoonlijke metingen van zwevend stof (PM10) zijn uitgevoerd bij 37 volwassenen in Amsterdam en bij 45 lagere school kinderen in zowel Amsterdam als Wageningen. Daarnaast zijn bij 13 kinderen uit Wageningen persoonlijke metingen van fijn stof gedaan. Metingen in de buitenlucht zijn gedaan op één vast meetpunt in zowel Amsterdam als Wageningen. Daarnaast zijn stofmetingen gedaan in de woonkamers van de volwassenen en in de klaslokalen van de kinderen. Er werd gestreefd naar 7-8 metingen per persoon. De middelingstijd van persoonlijke, buitenlucht- en woonkamer-metingen was 24

uur. In de klaslokalen zijn daarnaast ook 8-uurs metingen tijdens schooltijd gedaan. Met behulp van individuele lineaire regressie analyse is voor elk proefpersoon het verband tussen persoonlijke blootstelling enerzijds en buitenluchtconcentraties anderzijds berekend. Vervolgens is de verdeling van de individuele correlatie en regressie coëfficiënten onderzocht. Daarnaast is getracht om niveauverschillen tussen persoonlijke blootstelling en buitenluchtconcentraties te verklaren met behulp van met een vragenlijst verzamelde informatie over factoren die mogelijk de blootstelling aan stofvormige luchtverontreiniging kunnen beïnvloeden.

In hoofdstuk 2 worden de gebruikte methoden voor de persoonlijke PM10 en fijn stof metingen beschreven. De meetmethode is geëvalueerd door te kijken naar uitval van proefpersonen en metingen, aanzuigdebiet, weegprocedure, veldblanco waarden en door het uitvoeren van vergelijkingsmetingen tussen de persoonlijke meetmethode en stationaire meetmethoden. Bovendien is de mogelijkheid dat mensen hun gedrag veranderen als gevolg van het dragen van het persoonlijk monstername apparaat onderzocht door de tijdsbesteding op meetdagen te vergelijken met de tijdsbesteding op andere doordeweekse dagen. De uitval was laag; 95% van de proefpersonen die na de eerste meting hadden toegezegd om deel te blijven nemen, volbrachten de studie met succes. Op de eerste twee dagen van de fijn stofmetingen na, was 90% van alle metingen bruikbaar. Alle voor- en nametingen van het aanzuigdebiet waren binnen 10% van het vereiste debiet van 4 liter per minuut. De precisie van de weegprocedure was voldoende. Alle concentraties waren hoger dan de detectielimieten. Concentraties gemeten met de persoonlijke meetmethoden waren hoog gecorreleerd met ($R>0.90$) en niet aanmerkelijk verschillend van concentraties gemeten met stationaire meetmethoden. Volwassenen besteedden significant minder tijd buiten (0.5 uur) en meer tijd in huis (0.9 uur) op meetdagen in vergelijking met andere doordeweekse dagen. Voor kinderen werden geen significante verschillen in tijdsbesteding gevonden.

In hoofstuk 3 wordt de relatie beschreven tussen persoonlijke, binnenlucht en buitenlucht PM10 concentraties bij volwassenen. Het betrof 50 tot 70 jaar oude niet-rokende volwassenen, zonder rokende huisgenoten en zonder beroepsblootstelling aan stof. De mediane Pearson's correlatie coëfficiënt (R) tussen persoonlijke en buitenluchtconcentraties was 0.50. Uitsluiting van metingen waarbij de proefpersonen aan tabaksrook blootgesteld waren, verbeterde de correlatie naar een mediaan van 0.71. Schattingen van de

correlatie op groepsniveau gaven lagere R-waarden; 0.34 en 0.50, respectievelijk. Concentraties in de buitenlucht (gemiddeld 42 $\mu g/m^3$) waren hoger dan in de binnenlucht (gemiddeld 35 $\mu g/m^3$), maar lager dan de persoonlijke blootstelling (gemiddeld 62 $\mu g/m^3$). Het belangrijkste deel van het verschil tussen persoonlijke blootstelling en buitenluchtconcentraties kon worden toegeschreven aan blootstelling aan tabaksrook, het wonen aan een drukke weg en tijd doorgebracht in een voertuig.

Hoofdstuk 4 beschrijft de relatie tussen persoonlijke, klaslokaal- en buitenlucht-concentraties van PM10 gemeten bij 10 tot 12 jaar oude kinderen. De mediane R tussen persoonlijke en buitenluchtconcentraties was 0.63 voor kinderen met niet-rokende ouders en 0.59 voor kinderen met rokende ouders. Schattingen van de correlatie op groepsniveau gaven lagere R-waarden; 0.45 en 0.20, respectievelijk Het uitsluiten van dagen dat kinderen met niet-rokende ouders waren blootgesteld aan tabaksrook, verbeterde de R naar een mediaan van 0.73. De gemiddelde persoonlijke PM10 blootstelling was 105 $\mu g/m^3$; gemiddeld 67 $\mu g/m^3$ hoger dan de bijbehorende concentratie in de buitenlucht. Het belangrijkste deel van dit verschil kon worden toegeschreven aan blootstelling aan tabaksrook, hoge PM10 concentraties in de klaslokalen en lichamelijke activiteit binnen.

In hoofdstuk 5 wordt de relatie tussen persoonlijke en buitenlucht-metingen van fijn stof bij 10 tot 12 jaar oude kinderen beschreven. De mediane R was 0.86. De geschatte correlatie op groepsniveau was wederom lager (0.41). Persoonlijke fijn stof concentraties waren ook sterk gecorreleerd met de PM10 concentraties in de buitenlucht (mediane R 0.75). De persoonlijke blootstelling aan fijn stof was gemiddeld 11 $\mu g/m^3$ hoger dan de buitenluchtconcentratie. Na het uitsluiten van metingen waarbij kinderen aan tabaksrook waren blootgesteld was het verschil nog maar 5 $\mu g/m^3$. De fijn stof concentraties in het klaslokaal waren vergelijkbaar met en hoog gecorreleerd met de concentraties in de buitenlucht.

In hoofdstuk 6 worden de oorzaken van de hoge PM10 concentraties in de klaslokalen nader onderzocht. Door middel van XRF analyse is de elementsamenstelling bekeken van monsters van 11 dagen waarop metingen tegelijkertijd waren uitgevoerd in de 2 scholen in Amsterdam en in de buitenlucht. Voor de meeste elementen was de concentratie in de klaslokalen veel hoger dan in de buitenlucht, vooral tijdens schooltijd. De hoogste klaslokaal/buitenlucht ratios werden gevonden voor elementen die met name in

bodemstof voorkomen (Si, Ca en Ti). De enige elementen die niet verhoogd waren, waren elementen die met name afkomstig zijn van antropogene bronnen (S, Br en Pb) en van de zee (Cl). Voor S, Br en Pb, elementen die voornamelijk geassocieerd zijn met deeltjes kleiner dan 1 μm, werden bovendien significante correlaties gevonden tussen de klaslokalen onderling en tussen concentraties in de klaslokalen enerzijds en concentraties in de buitenlucht anderzijds. Voor de andere elementen waren deze correlaties in het algemeen laag. Deze resultaten geven aan dat de hoge PM10 concentraties in de onderzochte klaslokalen waarschijnlijk veroorzaakt zijn door resuspensie van grof stof en/of suspensie van bodemmateriaal.

Hoofstuk 7 bevat de algemene discussie van het onderzoek. In de eerste plaats worden de belangrijkste resultaten samengevat en vergeleken met de bevindingen van andere onderzoeken. Vervolgens worden de invloed van een aantal potentiële bronnen van vertekening en een aantal beperkingen met betrekking tot generaliseerbaarheid van het onderzoek besproken. Tot slot worden de implicaties en conclusies gegeven. De conclusie van het onderzoek is dat variaties in de tijd van de buitenluchtconcentraties van PM redelijk goed (PM10) tot zeer goed (fijn stof) correleren met de variatie in persoonlijke blootstelling. De lagere correlaties voor PM10 zijn waarschijnlijk het gevolg van de grotere invloed van (re)suspensie van grof stof op PM10 concentraties dan op fijn stof concentraties. De individuele correlaties waren hoger dan de correlaties op groepsniveau. Deze resultaten ondersteunen het gebruik van metingen op een vast meetpunt in de buitenlucht als blootstellingsmaat in epidemiologische tijdreeksstudies waarin de dag-tot-dag variatie in stofvormige luchtverontreiniging gerelateerd wordt aan de dag-tot-dag variatie in gezondheidseindpunten. De persoonlijke blootstelling aan stofvormige luchtverontreiniging was aanzienlijk hoger dan de concentratie in de buitenlucht, vooral voor PM10 bij kinderen. Deze niveauverschillen konden grotendeels worden toegeschreven aan blootstelling aan tabaksrook en (re)suspensie van grove deeltjes.

List of abbreviations

CC	Casella Cyclone
CI	Confidence Interval
CP	Coarse PM10 Particles (PM10 minus PM2.5)
CV	Coefficient of Variation
D_{50}	50% cutoff aerodynamic diameter
DL	Detection Limit
EPA	Environmental Protection Agency
ETS	Environmental Tobacco Smoke
FP	Fine Particles
HI	Harvard Impactor
PM	Particulate Matter air pollution
PM10	Particles with a 50% cutoff aerodynamic diameter of 10 μm
PM2.5	Particles with a 50% cutoff aerodynamic diameter of 2.5 μm
PTEAM	Particle Total Exposure Assessment Methodology
PS	Personal Sampler
R	Correlation coefficient
RSP	Respirable Suspended Particles
SA	Sierra Anderson
SAS	Statistical Analysis System
SD	Standard Deviation
SE	Standard Error
THEES	Total Human Environmental Exposure Study
TSP	Total Suspended Particulates
UL	Uncertainty Limit
XRF	X-Ray Fluorescence

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Curriculum Vitae

Nicole (Adriana Huberdina) Janssen werd geboren op 29 juli 1967 in Eindhoven. In 1985 behaalde zij het VWO diploma aan het van Maerlantlyceum te Eindhoven. Van 1985 tot 1992 studeerde zij Milieuhygiëne aan de Landbouwuniversiteit in Wageningen, met afstudeervakken bij de vakgroep Gezondheidsleer en de vakgroep Luchthygiëne en -verontreiniging. Na haar afstuderen werkte zij eerst een jaar als projectmedewerker aan verschillende onderzoeken bij de vakgroep Humane Epidemiologie en Gezondheidsleer. In april 1993 werd zij als assistent in opleiding (AIO) aangesteld bij de vakgroep Luchtkwaliteit. In het kader van deze aanstelling werd het in dit proefschrift beschreven onderzoek uitgevoerd. Sinds april 1997 werkt zij bij de afdeling Gezondheidsleer, aan een onderzoek naar de effecten van luchtverontreiniging van het weg- en luchtverkeer op de respiratoire gezondheid van kinderen.