Air pollution in Dutch homes

an exploratory study in environmental epidemiology

Erik Lebret

STELLINGEN

1.

Uitgaande van de gangbare definities, zoals gebruikt in de beschrijving van integrale persoonlijke blootstelling aan luchtverontreiniging, kan de woning meestal niet gezien worden als een 'micro-environment'.

(Dit proefschrift)

2.

Blootstellingsmaten voor luchtverontreiniging, gebaseerd op de aan- of afwezigheid van bronnen van NO_2 en respirabel stof in de woning, zijn in de regel onvoldoende nauwkeurig voor het gebruik in epidemiologisch onderzoek.

(Dit proefschrift)

3.

De observatie dat populaties in verschillende Nederlandse regio's verschillen in longfunctie parameters en CARA prevalentie, kan niet zonder meer verklaard worden door verschil in blootstelling aan luchtverontreiniging in de buitenlucht, aangezien er ook regionale verschillen kunnen zijn in blootstelling aan luchtverontreiniging in de woning.

Maas, P.J. van der (1979), CARA bij kinderen. Proefschrift Erasmus Universiteit, Rotterdam.
Lende, R. van der (1981), Bull Europ Physiopath. Resp. 17, 775-792.

4.

Aangezien er nauwelijks inzicht bestaat in het huidige ventilatiegedrag van bewoners, noch in de drijfveren daarvoor, en aangezien bovendien nauwelijks duidelijk is welk ventilatiegedrag wenselijk is, valt van een voorlichtingscampagne ter verbetering van ventilatiegedrag weinig effect te verwachten.

5.

The suggestion 'that all monitoring programs should be directed toward the formulation and validation of indoor air quality models, rather than toward the characterization of the (US) housing stock' is unjustified.

Moschandreas, D.J. (1982), J. Air Poll. Contr. Assoc. 32, 904-906.

6.

When reading standard textbooks on statistics or books on computers, one may easily gain the notion that a statistical analysis done on the computer is a fairly simple one-step operation; however, real life is not so simple, since each stage of the analysis uncovers information that affects the subsequent steps.

After: Dixon, W.J. ed. (1981), BMDP Statistical Software, University of California, Berkeley.

7.

De door Toffler beschreven 'demassificering' van de samenleving kan in de toekomst voor de milieu-epidemiologie tot gevolg hebben dat het retrospectieve en dwarsdoorsnede onderzoek naar oorzaken van chronische aandoeningen nog moeilijker wordt.

Toffler, A. (1980), The third wave. Pan Books Ltd., Londen.

8.

De besluitvorming rond problemen van verontreiniging in de woonomgeving zou kunnen verbeteren, indien, naast een risico-analyse, ook onderzoek naar de risico-perceptie van de betrokkenen zou worden uitgevoerd.

9.

In zijn evaluatie van het effect van een pakket thermische isolatiemaatregelen op de luchtinfiltratie in bestaande woningen, besteedt Meijer onvoldoende aandacht aan de mogelijkheid dat bewoners hun (ventilatie)-gedrag veranderd kunnen hebben nadat de isolatiemaatregelen zijn uitgevoerd.

Meyer, L.A. (1981), Energiebesparing in de sociale woningbouw; besparing op ruimteverwarming in theorie en praktijk. Proefschrift Rijksuniversiteit Groningen.

10.

Het verdient aanbeveling om in het onderzoek naar het zogenaamde 'sick building syndrome' meer aandacht te besteden aan wat Naisbitt omschreef als de 'high tech/high touch balance'.

Naisbitt, J. (1984), Megatrends. Warner Books Inc, New York.

11.

In de reclame voor afvoerloze petroleum-kachels dient de verwijzing naar een TNO-rapport, dat de onschadelijkheid van de verbrandingsgassen zou aantonen, vervangen te worden door de waarschuwing, dat deze kachels de NO_2-concentratie in de woning aanmerkelijk kunnen verhogen.

TNO Hoofdgroep Maatschappelijke Technologie, rapport nr. 8711-60654, Apeldoorn.

Stellingen behorend bij het proefschrift:
"Air pollution in Dutch homes; an exploratory study in environmental epidemiology".
Erik Lebret,
Wageningen, 2 oktober 1985.

Air pollution in Dutch homes

an exploratory study in environmental epidemiology

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Promotoren: dr. E. H. Adema, hoogleraar in de luchthygiëne en -verontreiniging

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Air pollution in Dutch homes

an exploratory study in environmental epidemiology

Proefschrift

ter verkrijging van de graad van
doctor in de landbouwwetenschappen,
op gezag van de rector magnificus,
dr. C. C. Oosterlee,
in het openbaar te verdedigen
op woensdag 2 oktober 1985
des namiddags te vier uur in de aula
van de Landbouwhogeschool te Wageningen

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STRUCTURE OF THIS REPORT.

The subject of air pollution in homes is of interest for people from various backgrounds. Medical doctors, environmentalists, building engineers, public health officers, gas industry and consumer organisations all have to deal with indoor air quality problems in one way or the other. It is therefore necessary to introduce briefly several issues of indoor air pollution, which may be commom knowledge for some readers but not for others.

The report is devided in two parts; one which describes the study and one which contains the results. The figures and tables are separated from the text.
In part 1, the subject of air pollution in homes and the background of the study are introduced in chapter 1. Chapter 2 gives an overview of indoor air pollution research methodology and sketches the current state of knowledge. In chapter 3, the study design is presented; this chaper is arranged according to the three main measurement programs which form the body of the study. The statistical techniques, which were used for the data analysis in part 2, are briefly introduced in the final part of chapter 3.

Part 2 presents the results of the study. First, a description is given of some general characteristics of the homes which were studied. In chapters 3 to 6, the measurement results for the pollutants under study are treated separately. In these chapters, the associations between indoor pollutant levels and characteristics of the homes are described and each chapter ends with a summary and discussion. In the final chapter, a general discussion on air pollution in Dutch homes is given from the perspective of public health.

"The equations are not reliable when applied to individuals, as you well know. They only deal with humanity in mass."

Isaac Asimov; Foundation's Edge.

1982

CHAPTER 1. INTRODUCTION

1.1. General introduction

Due to the "oil crisis" of 1973, the balance between the need for clean indoor air and the need for thermal comfort and protection against outdoor perils has been disturbed by increasing residential energy costs.The "oil crisis" led to governmental campaigns by which occupants were encouraged to reduce domestic energy consumption.
Activities like insulating homes and sealing up the building envelopes were subsidized by the Dutch government after 1973. Overly enthusiastic occupants even sealed ventilation ducts. More recently, there has been a tendency to change to other (and cheaper) heating systems, such as wood stoves and kerosene heaters. The activities to reduce energy consumption and expenses led to new sources of indoor pollutants and simultaneously the dilution of these pollutants was limited by the reduction of ventilation of the homes.

Parallel to this, new materials and new chemicals which were used for construction, wood conservation, home decoration and for the fabrication of a wide array of other consumer products were introduced into the homes. Concern that these developments might lead to an unacceptable deterioration of indoor air quality was expressed among others in the late Seventies in presentations at the Symposium on Indoor Climate, Copenhagen (Fanger and Valbjørn 1979) and at an international workshop "Health aspects related to indoor air quality", organized by the World Health Organization, Bilthoven (WHO 1979). In the same year, the Dutch Minister of Public Health and Environmental Protection addressed the Public Health Council with a request to advice him on minimal ventilation requirements in homes, needed for acceptable indoor air quality.

Although nonoccupational indoor air pollution goes back to the time when prehistoric man started to use open fires in enclosed spaces, the systematic research efforts in this field are from a rather recent date. In public health literature, air pollution has mainly been considered as an outdoor phenomenon. In the Dutch Law on Air Pollution (Anonymus 1973) air pollution is, by definition, explicitly restricted to the ambient atmosphere.
Besides domestic energy conservation and the introduction of new materials in the home, another factor has shifted attention from outdoor to indoor air pollution: the recognition of the existing time-activity patterns of people in western industrialized societies. Most people spend about 16 to

17 hours per day in their home (Szalai 1972, Chapin 1974, Knulst 1978). The actual exposure to outdoor air pollution is thus limited to short periods of time. Furthermore, the earlier studies on indoor air pollution have shown that indoor levels could be quite different from the outdoor levels, depending on type of pollutant and circumstances.
In most epidemiological studies in the past, population exposure estimates were based on ambient air pollution levels which were measured at one or a few fixed monitoring stations. In some cases, exposure estimates were based on whether people were living in an industrialized or a rural area. As a result of regulatory actions based on, among other factors, the findings of earlier health effect studies, the contrasts in ambient air pollution levels in time and place are now smaller than in the first half of this century with its more severe air pollution episodes. It is, therefore, questionable whether the use of traditional exposure estimates is still a valid approach in epidemiological studies, given the time use of people and the differences between outdoor and indoor air pollution levels.
In the Sixties, Biersteker and de Graaf(1967) were among the first to recognize the implications of indoor air pollution for health effect studies on air pollution. It is now generally acknowledged that obtaining accurate exposure measures is one of the most complicated tasks in the design of such epidemiological studies.

1.2. The concept of integrated personal exposure

Personal exposure to a pollutant can be defined as the event when a person comes into contact with a pollutant of a certain concentration during a certain time period (Ott 1980). while exposure to a particular pollutant may occur simultaneously through various pathways such as air, water, soil and food (Behar 1979), only exposure to air pollution will be discussed here.

The concentration of a pollutant at a certain location varies in time, and different locations have different concentrations. The definition of personal exposure implies that person and pollution are at the same place at the same time. People therefore encounter different concentrations of various duration at different locations, when moving from one location to the other during their normal daily activities. The durations (t) of the exposure of an individual (i) to a concentration (C) over a time period (ta), is taken into acount by the integrated personal exposure (E) (Bennet 1982, Ott 1980,1982):

$$E = {}_0\int^{ta} C\,(t)\,dt \qquad (\mu g.m^{-3}.h) \qquad (1)$$

or more conveniently, as time-weighed average,

$$\overline{E} = \frac{1}{ta} \cdot {}_0\int^{ta} C\,(t)\,dt \qquad (\mu g.m^{-3}, \text{ during period ta}) \qquad (2)$$

This definition does not imply any preference for any of the various ways in which exposure can be described in exposure-response relationships. From the perspective of possible health damage, other components of exposure to a pollutant, such as the magnitude of peak concentrations, intervals (recovery time) between peak exposures, duration of the exposure, etc., may be more appropriate to describe exposure-response relationships, depending on the pollutant and the effect under study.

Locations with a homogeneous pollutant concentration in space and time were called "microenvironments" by Ott (1980). Groups of similar locations were referred to as "microenvironment type" by Duan (1981), "location" by Fugas (1975) and "environmental mode" by Moschandreas and Morse (1979). In the following, the term "microenvironment" will be used.
For J different microenvironments, the integrated personal exposure can be formulated as:

$$E = \sum_{j=1}^{J} C_{ij} \cdot t_{ij} \qquad (\mu g.m^{-3}.h) \qquad (3)$$

or as time-weighed average,

$$\overline{E} = \sum_{j=1}^{J} C_{ij} \cdot t_{ij} \,/\, \Sigma\, t_{ij} \qquad (\mu g.m^{-3}, \text{during period } t_{ij}) \qquad (4)$$

where C_{ij} is the concentration in microenvironment (j) in which an individual (i) remained during a time period t_{ij}.

The integrated personal exposure can be assessed directly through measurements with personal monitoring devices, attached to persons during their normal daily activities.
In an indirect approach, integrated personal exposure can be calculated with formula (3) or (4), using data about the time people spend in different microenvironments and about the pollutant concentration in these microenvironments.Many different microenvironments (with dimensions of space and time) can be identified, but doing so limits the feasibility of

assessing time allocation and pollutant concentration with sufficient time resolution. Moschandreas and Morse (1979), for instance, have identified six different environmental modes (the occupational, the in-transit, the residential, the ambient, the office and the "others"), but some others have used up to 18 microenvironments.

Data from time budget surveys can be useful to estimate the time allocation of people in different microenvironments, with certain assumptions about the locations where the registered activities take place.
For the Netherlands, the average time allocation of 2730 people of 12 years and older was calculated from the original data of a time budget survey carried out in October and November 1980 (table 1).
Table 1 shows that, in the Netherlands, people spend about 90 % of their time indoors, and 70 % is spent at home. Just as in many other countries (National Research Council 1981), on the average only one to a few percent of the time is spent out of doors.
The time use of specific population groups may deviate substantially from the average time allocation of the general population. Farmers, for instance, will spend more time outdoors than average, while infants just as elderly and disabled people will spend more of their time in the home.

To assess the integrated personal exposure, research efforts must be directed to critical microenvironments. Critical microenvironments are those that have a large contribution to the integrated personal exposure. From formula (3) it can be seen that microenvironments with a high value for the product of pollutant concentration with time allocation can be considered as critical microenvironments.
With the figures of time use in mind, it is evident that, in general, outdoor air pollution can only have a substantial contribution to the integrated personal exposure when outdoor pollutant levels are similar to, or many times higher than those encountered in buildings.

1.3. Background and objective of the study

In 1980, a study was initiated regarding the integrated personal exposure of people to air pollution, with special reference to the possible health effects of the exposure. The investigation was conducted by the Department of Environmental and Tropical Health and the Department of Air Pollution of the Agricultural University Wageningen and was began with a literature survey on indoor air pollution and public health (Boleij and Brunekreef 1982).

The subject of this thesis is the first step in the staged assessment of personal exposure.
The objective of this first step was the characterization of the typical pollutant levels inside Dutch homes, in relation to properties of the home and occupants, from a perspective of integrated personal exposure and public health.

The characterization of all known indoor air pollutants would be beyond the scope of the study. Therefore, the study was initially restricted to:

- carbon monoxide (CO)
- nitrogen dioxide (NO_2)
- respirable suspended particles (RSP)

The selection of these general air pollutants was based on the following considerations.
First, these pollutants have known indoor and outdoor sources in the Netherlands. Second, these pollutants are generated by a limited number of indoor sources. Third, health effects of these pollutants have been documented. In particular, indoor exposure to NO_2 and RSP is suspected of playing a (yet not fully understood) role in the aetiology of chronic non-specific lung disease(CNSLD). These pollutants were also recommended for further study by Working Groups on Indoor Air Pollution of the WHO (WHO 1979a,1982).

Later, the study was extended to Volatile Organic Compounds (VOC). The interest in VOC stems from the recent discoveries that chemical waste in the soil of urban areas may act as a source of indoor air pollution and especially of VOC. The determination of VOC levels in Dutch homes was carried out in cooperation with the National Institute for Public Health and Environmental Hygiene, Bilthoven. In the case of VOC, the study objective was to generate a data base of typical indoor levels of VOC, against which levels in homes built on polluted soil could be evaluated.

Additional funds for the study were granted by the Ministry of Housing, Physical Planning and Environment.

1.4. Brief description of the pollutants under study

Carbon monoxide

Carbon monoxide (CO) is a gas generated by incomplete combustion of fossil fuels. CO inhibits the transport of oxygen in the body by forming carboxy-hemoglobin (COHb) in the blood. Its affinity to red blood cell hemoglobin

is about 240 times stronger than that of oxygen. Elevated carboxyhemoglobin levels can cause symtoms such as headache, dizziness, nausea and also an increased risk of angina pectoris in people with coronary artery disease. Very high levels cause asphyxia and death (WHO 1979b).
The Dutch Health Council (1975) has recommended maximum values for ambient carbon monoxide: 40 mg/m^3 (1-hour average) is not to be exceeded more than once a week; 10 mg/m^3 (8-hour average) is not to be exceeded more than once a month. With CO concentrations below these guidelines, the carboxyhemoglobin level stays below 2.5-3.0 %, a level considered safe for the general population, including sensitive individuals. In the period from April 1981 to March 1983 these guideline values were not exceeded by ambient concentrations measured at the stations of the National Monitoring Network (RIV 1981, RIV 1982). In the Netherlands, 1-hour average ambient CO levels of less than 5 mg/m^3 are typical.

Nitrogen dioxide

Nitrogen dioxide (NO_2) is formed in combination with NO, from atmospheric nitrogen and oxygen in high temperature combustion processes. From the point of view of health effects, NO is relatively harmless. NO_2, however, can denaturate the chemical structures in pulmonary tissues due to its oxidizing properties. Furthermore, there are indications from animal experiments that NO_2 can reduce resistance against airway infections. Experiments with healthy human volunteers have shown that exposure to NO_2 can increase the airway resistance and can increase the sensitivity to stimuli which lead to broncho-constriction (WHO 1977). In short, NO_2 is suspected to cause, aggravate, or maintain respiratory disease.

The proposed Dutch air quality standard for outdoor NO_2 are aimed especially at preventing short-term peak concentrations (Dutch Health Council 1979): a value of 150 $\mu g/m^3$ (24-hour average) is not to be exceeded more than one day per year; the 1-hour average value, not to be exceeded more than once a year, is 300 $\mu g/m^3$.
In the period from April 1981 to March 1983, 24-hour average ambient concentrations of about 30 $\mu g/m^3$ were typical in the Netherlands (CBS 1982, CBS 1983). Occasionally, the proposed standard was exceeded at monitoring stations near roads with a high traffic density (RIV 1982).

Respirable suspended particles

Respirable suspended particulate matter (RSP) is the fraction of airborne particles with an aerodynamic size of < 10 µm and, depending on definition,

a 50 % cut-off at an aerodynamic size of 2.5 to 5 μm (ISO 1981). RSP is the size fraction which can penetrate into the lower airways.
Various effects of RSP on human health have been described: chronic non-specific lung disease, mutagenic and carcinogenic effects and specific toxicological effects (e.g. for lead). Much of the effects of RSP depend on physical structure and chemical properties.
The standard for outdoor particulate matter in the Netherlands is only applicable for 'Standard Smoke' measurements and is related to ambient SO_2 levels. According to the US primary standard for outdoor particulate matter, a 24-hour average concentration of total suspended particulate matter (TSP) of 260 $\mu g/m^3$ should not be exceeded, while the annual geometric mean TSP level should not exceed 75 $\mu g/m^3$. In a revision of the standard it is proposed to replace TSP by an indicator for particles of less than 10 μm(PM_{10}). The proposed 24-hour average PM_{10}-limit ranges between 150-250 $\mu g/m^3$, while the annual arithmic mean PM_{10}-limit ranges from 50-60 $\mu g/m^3$ (EPA 1984).
Typical outdoor TSP levels in the Netherlands are between 40-90 $\mu g/m^3$ (24-hours average) with occasionalvalues up to 200 $\mu g/m^3$ (CBS 1982, 1983).

Volatile organic compounds

The term volatile organic compounds (VOC) refers to a wide range of air contaminants, from the lower weight, straight-chain saturated hydrocarbons to more complicated structures as, for instance, chlorinated aromatic compounds. An equally wide range of toxicological effects is described for VOC or their metabolites. For the most part, the reported effects are derived from animal experiments, or from investigations of occupational exposures. Among others, irritation of skin and mucous membranes, liver and kidney damage and carcinogenic and neurotoxic effects have been found for selected VOC (National Research Council 1981).
Ambient concentrations of most individual VOC remain below 5 $\mu g/m^3$. For toluene and xylenes ambient levels of a few tens of $\mu g/m^3$ are typical, but in areas with high traffic density, levels of over 100 $\mu g/^3$ have been found. The ambient levels of VOC are usually orders of magnitude lower than occupational standards.

CHAPTER 2. INDOOR AIR POLLUTION RESEARCH; AN OVERVIEW

2.1. Introduction

Probably the first person who studied an indoor air pollution problem in the Netherlands was Fokker (1884). In public gathering places, he exposed rabbits to air polluted with tobacco smoke and determined CO-levels in the blood of the animals. Since the mid-sixties, indoor air pollution has received an increased attention and more sophisticated measurement techniques were adopted. In 1972, Benson et al. compiled a literature review which included 77 references of mostly recently published work. In the following years several hundreds of publications with relevance to some aspects of the field of indoor air pollution have been issued (National Research Council 1981).

In general, the fast growing body of literature on indoor air pollution is compiled from different disciplines. The different perspectives of, for instance, the gas industry, ventilation engineering, energy conservation and public health protection have led to a very fragmentary character of the literature, since research objective, instrumentation and study design were (in part) determined by the background of the research institute or sponsor. This hinders a systematic assessment of the literature and makes it difficult to compare the results of the different investigations.

In this chapter, the indoor air quality research will be classified into three main approaches, each with a different emphasis on the factors influencing indoor air pollution. After these three approaches have been outlined, the current state of knowledge will be summarized for the pollutants under study.

2.2. Indoor air pollution research methodology

Research on indoor air quality can be classified in a mechanistic, case study and survey approach, depending on the emphasis that is put on different aspects of the factors that control indoor air quality, and the scale of the investigation. The factors that influence the pollutant levels within a structure are the air quality outside the structure, air exchange with the exterior, pollutant production within the structure, the geometrical properties of the structure and the removal of pollutant by chemical reactions, deposition, etc..

lance equation (5) is often used to describe the process of generation and elimination of indoor air pollution (National Research Council 1981).

$$V \frac{dCi}{dt} = P - E - Q\,(Ci - Co) \qquad (5)$$

V = volume of the structure (m^3)
Ci = pollutant concentration in the structure ($\mu g/m^3$)
P = emission rate of the pollutant inside the structure ($\mu g/h$)
E = elimination rate of the pollutant through chemical and physical processes ($\mu g/h$)
Q = air exchange rate with the exterior (m^3/h)
Co = pollutant concentration outside the structure ($\mu g/m^3$)
t = time (h)

This model applies to a single well-mixed compartment; more elaborate models contain factors for mixing, recirculation and filtering, or relate to multicompartment situations.

In the mechanistic approach, the investigations are carried out in the laboratory, experimental room, or in normal homes under controlled experimental conditions. Emphasis is put on determination of pollutant source strength, decay, the effect of ventilation and infiltration and/or on modelling of the whole process of pollutant production and elimination; the influence of occupant behaviour is excluded (Cote et al. 1974, Dewerth and Himmel 1974, Shair 1974, Hollowell et al. 1976, Moschandreas et al. 1978, Alzona et al. 1979, Hollowell et al. 1979). Research of this type addresses the indoor air pollution potential. It answers questions such as:
- to what degree is being indoors a protection against ambient air pollution?
- what is the CO- or NO_2-production of a cooking range or cigarette?
- what effect will reduction of ventilation (to conserve energy) have on indoor air quality?

In the case study approach, pollutant levels in one or a few occupied homes are monitored in detail over several days or weeks under normal living conditions (Cote et al. 1974, Moschandreas et al. 1978, Stevenson et al. 1979). Emphasis is put on determination of (the variability of) indoor pollutant levels (the term $\frac{dCi}{dt}$ of the mass balance(5)).

These studies supply information about the transient concentration patterns, distribution of pollutants within the home and about indoor/outdoor relationships. The studies involve bulky and noisy equipment which is

more commonly used in ambient air pollution research; the equipment is mostly placed outside the homes in mobile air pollution research laboratories. The case study approach is a verification under real life conditions of the indoor air pollution potentials derived from mechanistical studies. The differences in instrumentation and study design between mechanistic and case studies, however, are not always distinct.

The survey approach is often used in conjunction with epidemiological studies; pollutant levels are determined in a number of homes of (subgroups of) participants in a health effect study (Biersteker 1966, Speizer et al. 1980, Goldstein et al. 1979, Keller et al. 1979).
The purpose of these studies is to provide information about the distribution of exposure to indoor pollutants among participants, and to yield or verify simple predictive exposure estimates based on simple home characteristics.

The survey approach has received increased attention in recent years due to the development of instrumentation more suitable for measurements inside homes than the traditional ambient air pollution instrumentation used in the mechanistic and case study approach. In particular, the advances in personal monitoring devices contributed to this development.

2.3. State of knowledge on air pollution in homes

Separately, the usefulness of the three approaches to make inferences about pollutant levels in the general housing stock is fairly limited. The mechanistic and case study approach supply detailed information about the effect of the factors which influence indoor air pollution, but often yield results with a site-specific character.
The applicability of laboratory derived emission rates of gas appliances to field situations can be questioned (Traynor et al. 1983), just as the applicability of the standard usage patterns of these appliances developed by the gas industries is questionable (Dijkhof and Ogink 1978, Welch et al. 1982). Sterling and Kobayashi (1981) for instance, reported that in 11 % of 119 dwellings in a poor neighbourhood in New York, the gas range is used continuously for supplementary space heating, a substantial departure from the average range operation time of 1.5 hours reported by Welch et al. (1982). The dilution and exfiltration of indoor pollutants and the infiltration and ventilation of occupied homes is also poorly understood. Anecdotical reports show the complexity of the matter: depending on the use of windows, the air exchange rate of a single room may vary between 1 to 20 h^{-1} (Phaff et al. 1980); in homes with thermal insulation the doors between

rooms are left open more frequently and ventilation provisions are used more often than in similar homes without thermal insulation (van Raay and Verhallen 1980).

The survey approach supplies better information about the distribution of indoor pollutant levels among homes than the mechanistic and case study approach. However, since the distribution of the factors which influence indoor air pollution is often only poorly described in the survey type studies, the applicability of the results to other groups of homes is largely unknown. Moreover, little information is available about the distribution of these factors over the existing housing stock.

A general description of the current state of knowledge on air pollution inside homes is summarized in table 2.

A systematic appraisal of indoor air pollution in the general housing stock has not been attempted to date to our knowledge. It is generally acknowledged that this is one of the most serious gaps in our present knowledge, but also the most difficult to bridge. (National Research Council 1981, WHO 1982).

2.4. Sources and pollutant levels inside homes

Several indoor sources have been identified for the pollutants under study, the main groups being fossil-fueled combustion processes, tobacco smoking, building materials and consumer products. For reasons discussed earlier, it is difficult to compare the results of the various types of investigations and to discuss them separately. Instead, a compilation of results is presented in table 3.

The wide range in indoor concentrations in the presence of indoor sources is apparent in table 3. In general, the measurements under experimental conditions yielded somewhat higher pollutant levels than those under normal living conditions. The highest concentrations in table 3, however, are not the result of unrealistic experimental conditions, but have been found in normal occupied homes.

Carbon monoxide

Carbon monoxide is produced indoors mainly by unvented gas-fired cooking ranges, waterheaters, by gas- or kerosene-fired space heaters and woodfires

(Elkins et al. 1974, Wade et al. 1975, Bartholomeus and Kaijser 1976, Hollowell et al. 1976, Sterling and Sterling 1979, Stevenson et al. 1979, Moschandreas et al. 1980). CO can also be emitted indoors by tobacco smoking (Corn 1974, Harke et al. 1974, Grandjean et al. 1979). Space heaters with faulty flues, automobiles in basement garages, wood stoves and cloth dryers have also been reported as occasional indoor sources of CO (National Research Council 1981). Most of these investigations on indoor CO had a mechanistic or case study character. The results indicate that, in the absence of indoor sources, the indoor CO concentration equals outdoor levels.
Tobacco smoking can elevate indoor levels by a few mg/m^3; occupants will in general keep CO levels from smoking below 10 mg/m^3, to avoid the irritating effect of other smoke constituents. Gas appliances and other combustion processes can lead to levels of a few tens of mg/m^3, and peak levels of over 100 mg/m^3 (cf. above references).

Nitrogen dioxide

Nitrogen dioxide is emitted indoors by the same unvented, fossil-fueled combustion appliances that produce CO. It was, therefore, often subject of investigation in the same studies as CO. Since the Palmes diffusion tube (Palmes et al. 1976), a passive sampler for NO_2, became available, survey type data for NO_2 have been gathered in several studies in recent years.
In the absence of indoor sources, indoor concentrations are 20-60 % lower than outdoors, due to the reactivity of NO_2. In homes with gas ranges and unvented gas or kerosene heaters, indoor levels of several tens, sometimes hundreds of μ^3, are typical. Peak levels of over 1000 $\mu g/m^3$ have been observed (Cote et al 1974, Elkins et al. 1974, Hollowell et al. 1976, Goldstein et al. 1979, Keller et al. 1979, Moschandreas et al. 1979, Palmes et al. 1979, Speizer et al. 1980, Stevenson et al. 1979).

The impact of tobacco smoking on indoor NO_2 levels is not well established, but is probably small compared to unvented fossil-fueled combustion appliances. Good et al. (1982) found NO_2 levels in homes with smokers only a few $\mu g/m^3$ higher than in homes without smokers.

Respirable suspended particles

The earlier studies on indoor suspended particles involved the total suspended particle concentration, instead of the respirable fraction of particulate matter. General conclusions of earlier studies were (Yocom 1982):

- indoor suspended particulate matter has another size distribution and composition than outdoor particulate matter,
- indoor/outdoor concentration ratios are less than one for the larger particles due to filtering; the smaller particles have a ratio of about one in the absence of smokers, but much higher in the homes of smokers.

For RSP the best documented and probably predominant indoor source is tobacco smoking. Other reported sources are resuspension, condensation of vapors, aerosol sprays, hobby and homecraft activities and combustion products. Most of the data on indoor RSP levels are of the mechanistic or survey type.
In the absence of smokers, indoor RSP levels are comparable with outdoor levels; for smoker's homes, levels of several tens, sometimes hundreds of $\mu g/m^3$ have been reported (Moschandreas et al. 1978, Spengler et al. 1979, 1980, Repace and Lowrey 1980, Diemel et al. 1981).

Volatile organic compounds

VOC have numerous indoor sources including bioeffluents, building, furniture and decoration materials; furthermore they are emitted indoors as solvent, propellant or active ingredient of a wide array of consumer products. Most of these sources are only (poorly) documented in a few mechanistic or case studies (National Research Council 1981). Virtually all studies indicate that many of the VOC have indoor/outdoor concentration ratios considerably higher than one.

2.5. Personal exposure studies

A special group of investigations related to the field of indoor air pollution is formed by the personal monitoring studies. In this type of study (mostly of recent date) the integrated personal exposure to a pollutant is assessed with personal monitors, and compared with pollutant levels at fixed monitoring stations (including the home), which represent different microenvironments.
From these studies inferences can be made about the effect of the home environment on personal exposure and about the bias of exposure estimates based on ambient pollutant levels.

Ziskind et al. (1982) found for their 10 subjects that on workdays the home environment contributed 18 % to the personal exposure to CO, against 40 % for the work environment and 23 % for commuting. On weekend days these

percentages were 40, 4 and 10 respectively. These results corroborate the findings of Cortese (1976).
Fugas et al. (1982) observed that personal exposure of their 12 volunteers to CO as well as RSP in the winter period, agreed best with the exposure at home, but poorly with exposure to outdoor concentrations.
The impact of indoor concentrations on exposure to RSP was also reported by Spengler et al. (1980). Personal exposure of 37 participants was determined in part by outdoor levels, but indoor concentration and passive exposure to tobacco smoke were much better estimates for personal exposure.
It has also been demonstrated that the indoor NO_2 levels have a substantial contribution to personal exposure (Dockery et al. 1980, Nitta and Maeda 1982, Quackenboss et al. 1982). Dockery and co-workers concluded that personal exposure to NO_2 could be adequately predicted from the ambient NO_2 level and the type of cooking fuel in the home.
In summary, the personal exposure studies have demonstrated that a substantial amount of the variance in personal exposure to air pollutants can be explained by the presence or absence of pollutant sources (cooking fuel, tobacco smoking) in the home.
Several epidemiological studies have adopted the approach of using the presence or absence of indoor pollutant sources as an estimate for personal exposure (Melia et al 1977, 1979, Keller et al. 1979, Comstock et al. 1981, Hasselblad et al. 1981, Kauffmann et al. 1983, Tager et al. 1983).

2.6. Indoor air quality in the Dutch housing stock

2.6.1. The housing stock

At the time of the start of the study, there were over 4.5 million homes in the Netherlands. A register of all Dutch homes is maintained by the Central Bureau of Statistics. This register is a compilation of the National Census data of 1971 and data of the homes which were built after 1971. However, buildings which have either lost their housing function or are demolished, are not removed from the register. Data aggregated over the whole housing stock are not readily available from the register.

The most recent and complex overview of the condition of the general housing stock is provided by a survey on the quality of the housing stock carried out in over 1800 homes in about 100 municipalities (KWO 1977). The main objective of this study was to compile information about the character and structure of the housing stock, in relation to the quality of technical and housing aspects. The study has supplied a wealth of information on the

distribution of age, ownership, rent rates and several building characteristics of the housing stock. Yet, the study results do not offer enough detail about the distribution of factors controlling indoor air pollution, to make inferences about indoor air quality in Dutch homes, or to select groups of homes that may be of special interest from an indoor air quality perspective.

2.6.2. Indoor air quality regulations

There are several regulations in the Netherlands which are pertinent to indoor air quality. For the pollutants under study the most important are:
- NEN 1087 (1975) Ventilation of homes;Requirements.
- NEN 1078 (1978) Installation code for natural-gas fired appliances.

In addition, the NPR 1088 (1975) Ventilation of homes;Practical Guidelines (1975) and the Model Building Regulations (VNG 1982) provide guidelines and examples for construction details to meet the requirements of NEN 1087 and NEN 1078. Other regulations such as the Commodities Act, Pesticide Act, Housing Act and in future the Chemical Substances Act provide a potential framework for future regulatory actions.

The ventilation standard for homes (NEN 1087) is based on the need for a fresh air supply of 25 m^3 per hour per person per room, from the perspective of human physiology. Ventilation requirements for different types of rooms according to NEN 1087 are given in table 4.
The air supplied to rooms should be sufficiently pure; it is implied that this supplied air should not been drawn from other rooms.

The installation code for gas appliances (NEN 1078), prohibits the use of unvented gas appliances in homes. For cooking ranges, ovens and water heaters, exceptions to this rule are made under certain conditions. The code only applies to gas appliances installed after 1978; older appliances have to comply with older versions of the code. The part of the code which covers ventilation and exhaust of combustion products, aims at maintaining proper air quality needed for human safety and proper functioning of the gas appliances.
Carbon monoxide concentrations higher than 58 mg/m^3 (50 ppm) and carbon dioxide concentrations higher than 3 % (3000 ppm) are not allowed; CO_2 levels of less than 1% are considered desirable.
Combustion products other than CO, CO_2 and water vapor (such as nitrogen oxides) are not considered in NEN 1978.
The code specifies the minimal air supply for specific situations. In addition, it specifies the permanent ventilation provisions which are

needed to attain this air supply.
For vented appliances the required ventilation is 2-4 m^3 air per hour per kWatt energy capacity, depending on the ratio of the room volume to the capacity of the appliances; for unvented appliances a minimum of 9 m^3 air per hour per kWatt energy capacity is required.

2.6.3. Current state of knowledge on indoor air pollution in the Netherlands

In the past, only two of the pollutants under study (CO and RSP) have been subject of investigation in Dutch homes.
In 1973/1974 the CO production of natural gas-fired appliances was determined in a field study. The purpose of the study was to compile information about the condition of domestic gas installations and maintenance practice. Among the investigated appliances were 1200 'geisers'. The 'geiser' is an instantaneous water heater which is usually located in the kitchen and supplies the home with hot water and often the shower as well. At present, they are used in 70 % of the Dutch homes and are usually unvented.
In this study the so called 'toxicity index' (the ratio of CO to CO_2) in the flue gases of the geiser was determined. The installation code for gas appliances specifies a maximum toxicity index of 0.01, which is roughly equivalent with about 600 mg/m^3 CO (36 mg/MJ or 28 mg/min) in the flue gases. Eleven percent of the tested geisers had CO emission rates which exceeded these values; 8 % had a toxicity index of 0.02 or higher. Reasons for the high emission rates were soiling of the burner and lack of maintenance of the appliances (De Vries and Bartholomeus 1973, Bartholomeus and Kayser 1976, Vermünicht et al. 1979).
Borst (1972) carried out an investigation in the city of Enschede in 237 homes equipped with a geiser. In 16 % of the kitchens, the CO level was higher than 285 mg/m^3 (250 ppm) after 30 minutes of operation of the geiser under standardized conditions.
The results of this study prompted the implementation of a stricter maintenance system for gas appliances in this city.

The lethal effect of indoor CO pollution from geisers is illustrated by statistics of CO poisonings in table 5.
In the Sixties, town gas was replaced by natural gas in the Netherlands and coal was rapidly abandoned as a home heating fuel. The statistics on CO poisoning reflect these changes. CO poisoning decreased dramatically due to the abandoning of coal, but at the same time the geiser emerged as a new source for CO. The geiser-related fatalities have in common that several factors such as poor ventilation, prolonged appliance use, inadequate

flues, etc., act concomitantly, which leads to exceptional high CO concentrations.To what extent lower, but still elevated, indoor CO levels impose a health risk is not clear.

Suspended particulate matter (in combination with SO_2) was measured by Biersteker and co-workers (1965) in Rotterdam. At 65 homes, 'standard smoke' samples (a method to determine the concentration of suspended particulate matter) were taken indoors and outdoors. On average, the indoor standard smoke concentration was about 80 % of the outdoor standard smoke concentration. The indoor standard smoke concentration was positively associated with indoor tobacco consumption.

Long-term average concentrations of respirable suspended particles were determined in the Netherlands in an epidemiological study in 84 homes near a secondary lead smelter (Diemel et al 1981, Brunekreef and Boleij 1982). Most of the homes were built between World War I and II, were small in size and were mainly occupied by people of relatively low educational and occupational status. In two periods of one month each, the average concentration of RSP ranged from 20 to 570 $\mu g/m^3$. Compared to levels reported for indoor RSP in other countries, the concentrations were high. Levels of the first and second period were highly correlated. In 26 homes for which information about the number of smokers was available, a clear relation was found between indoor RSP level and the number of smokers in the home.

In summary, the current knowledge about air pollution in Dutch homes is scant. For CO, the potentially high production of geisers was established; suspended particulate levels were found to be clearly related to tobacco smoking, and long term average concentrations in 84 homes were high compared to the levels reported for other countries.

2.6.4. Distribution of indoor pollutant sources in the Dutch housing stock

For lack of other information, more general information about the distribution of known indoor pollutant sources in the Dutch housing stock had to be used for a first evaluation of indoor air quality in the Netherlands.
One group of sources which produce CO, NO_2 and to a lesser extent RSP, is the group of the fossil-fueled combustion appliances.
Due to the large resources of natural gas in the Netherlands, 90 % of the homes is connected to the gas network and supplied with natural gas (Hemrica 1980). In about 85 % of the homes, a gas-fired cooking range is in use and about 70 % of the homes is equipped with a gas-geiser as water heater. Of these geisers, 18 % receives no regular maintenance, which makes the

burners liable to soiling and therefore to incomplete combustion (Boonstra and Zwetsloot 1981). Open wood stoves are found in 14 % of the (mostly private and upperclass) homes. Unvented natural-gas fired space heaters are not allowed in the Netherlands by the gas installation code.
Recently, unvented kerosene-fired space heaters have been introduced in the Netherlands, but they are not widespread yet.

Smoking was one of the other identified sources of indoor air pollution. In 1979, 52 % of the Dutch males and 38 % of the females of 15 years and older were smokers. On average the males smoked 15 cigarettes/day, against 13 cigarettes/day smoked by the females. Over the years, there is a decrease in the total number of smokers, but an increase in the total amount of tobacco that is consumed (De Haas and De Haas 1981).

Little information is available about the distribution of indoor VOC sources in the housing stock, but it is evident that in most homes several of the recognized indoor sources are present.
With the present state of knowledge it is impossible either to translate the results from the few Dutch studies to the general housing stock or to make inferences from the foreign literature.
The widespread use of notorious indoor pollutant sources in Dutch homes, however, warrants the concern that for the pollutants under study, the indoor levels may be higher than ambient levels. The questions to what extent the indoor sources affect indoor levels in Dutch homes, how this reflects the personal exposure of the occupants and whether this indoor pollutant exposure may have negative effects on public health cannot be answered without further study. In the next chapter, the design of the study which addresses the first of these questions is discussed.

CHAPTER 3. STUDY DESIGN

3.1. Introduction

The aim of the study was:
- the characterization of the distribution of pollutant levels inside Dutch homes,
- the identification of the factors which influence indoor pollutant levels, and
- the development and evaluation of simple predictive models to estimate human exposure to indoor pollution, for use in epidemiological studies.

The public health perspective of the study had several consequences for the study design and instrumentation.
First, the characterization of indoor air pollution should cover a wide range of different (types of) homes under normal living conditions, to assess the exposure of the general population to indoor pollutants. Given the diversity of the housing stock a survey approach would therefore be necessary.
Second, the exposure to a pollutant is not very relevant when seen separately from the complicated mechanisms through which adverse health effects occur (although these mechanisms are often only poorly understood). Therefore information was needed about the magnitude of peak concentrations, the intervals between peak concentrations and the consistency of indoor pollutant levels over longer periods of time. This called for a more mechanistic study approach.

Elements of both the mechanistic and the survey approach were combined in three related measurement programs which together form the body of the study.In addition to the three main measurement programs, several smaller studies on specific aspects of indoor air pollution were carried out within the framework of the study, often as graduate work of students. The latter studies had the character of pilot studies to assess the feasibility of different measurement methods and epidemiological study designs. When appropriate, the results of these studies will be presented as far as they are relevant to the characterization of indoor air pollution.

3.2. General outline of the three main measurement programs

The three main measurement programs were:
- real-time monitoring of pollutant levels in different rooms of a limited number of homes,
- week-long integrated measurements in a large number of homes, and
- repeated week-long integrated measurements of pollutant levels over different seasons in a small set of homes.

The first two programs were carried out in the heating season (± October - March), when the ventilation of homes was expected to be at a minimum.

The purpose of the real-time monitoring program was to determine the transient concentration peaks of CO and NO_2 in relation to the use of unvented gas appliances, the dispersion of pollutants through the home, and the day to day variation in pollutant levels over a period of about 7 days. For RSP and VOC no real-time continuous monitors were available. Tracer gas experiments using sulphurhexafluoride (SF_6) were incorporated in both the real-time and the week-long integrated measurement program. The tracer gas experiments are described in appendix A; the objective of the tracer gas experiments was the determination of kitchen ventilation, dispersion of kitchen air to the living room, and the determination of the effect of kitchen ventilation on indoor pollutant levels.

The objective of the week-long integrated measurement program was to determine the distribution of pollutant levels for all the pollutants under study in a wide range of homes typical for the housing stock, in relation to characteristics of the home and its occupants.
A measurement period of one week was chosen because it was thought that many occupant activities which affect indoor pollutant levels (cooking, smoking, ventilation of rooms, etc.) are carried out in 24-hour cycles, with the exception of weekend days, which have their own rhythm (shopping, recreation). A week-long measurement was therefore considered optimal to cover much of the regular household activities. Shorter periods would be more affected by chance variation due to incidental irregular activities, while longer periods would reduce the number of homes to be included in this program, due to logistic restrictions.

The repeated measurements were intended to evaluate the variability of indoor pollutant levels within and between different seasons. This would give an impression of the number of measurements necessary to characterize pollutant levels in a home, and of the errors associated with the use of a single indoor measurement to estimate exposure to indoor pollutants.

3.3. Real-time monitoring program; material and methods

Instrumentation

The real-time monitoring program was performed with conventional air pollution monitors: a dual range Ecolyzer 2000 to monitor CO-levels, a Bendix chemiluminescence NO_x monitor and a Wilks infrared spectrophotometer to determine SF_6 tracer gas concentrations (see figure 1).

Samples for CO and NO_x monitors were drawn from 3 indoor locations (kitchen, living room and bedroom) and 1 outdoor location through 15 meter 1/8 inch Polyflow sampling lines, using Metal Bellow (model MB-21E) pumps. A 5-way valve (Hoke type 7941 G 4 Y) with a Ramcon 3B-WP actuator was used to switch from one sampling location to the other. Thermo couples were used to monitor the use of cooking range and geiser. The infrared spectrometer sampled only kitchen air.
Data acquisition and control of the actuator was handled by a Hewlett Packard model 85 micro computer. The micro computer was programmed for a sampling sequence which first allowed the sampling lines to be flushed for one minute (more than twice the systems response time), then the readings of monitors and thermo couples were integrated over the following minute and stored on tape. After this, the 5-way valve was activated to switch to the next location. The indoor locations were sampled 8 or 9 times each hour. Outdoor samples were taken only once every hour to achieve a more frequent sampling of indoor locations.

SF_6 tracer gas was emitted in the kitchen at a constant flow of about 1 ml/min from a 1 liter stainless steel cylinder with a Brooks flow controller (model 8844; needle size 2). Thus a constant SF_6 emission could be maintained over a period of over 7 days. Between homes, the SF_6 emission varied from 165-300 mg/hour.

Calibration equipment and procedure

The field calibration of the CO monitor was carried out with an electronic zero signal (and adjustment) and a 12 mg/m^3 CO span gas. The CO span gas was introduced into the monitor from a 10 liter Linde Plastigas bag filled from a calibration cylinder at the laboratory. Periodically the interference filter of the monitor was checked on proper functioning.

The field calibration equipment for the NO_x monitor consisted of a cylinder with zero air, a certified cylinder Spectraseal NO calibration gas of 195

$\mu g/m^3$, and a Tracor portable permeation chamber with a NO_2 permeation tube. After zero adjustment with zero air, the NO and NO_x signals of the monitor were calibrated against the NO span gas. Then the permeation device was flushed with zero air from the cylinder, which resulted in a NO_2 span gas concentration between 750-1500 $\mu g/m^3$ with traces of NO. This gas was introduced into the monitor and the NO_2 signal was adjusted against the NO_x reading after subtracting the NO concentration ($NO_2 = NO_x - NO$).
Periodically the NO_x monitor was checked against a permanent NO_2 dynamic dilution system and compared with other monitors at the laboratory.

The infrared spectrophotometer was calibrated according to the operation manual. Cell path length was set at a maximum of 25 meter and the wavelength at 11.6 μm. The measurement cell was filled with zero air and closed. After zero adjustment, the cell inlet and outlet were short circuited and known volumes of pure SF_6 from a Plastigas bag were injected through a septum into the known cell volume; in this way, scale readings of at least 3 different concentrations in the range from 0-12 mg/m^3 SF_6 were obtained.
Periodically, checks were made by introducing calibration gases of various concentrations into the cell from cylinders prepared at the laboratory.

Selection of homes

People in the neighbourhood of Wageningen were invited to participate in the real-time measurement program by means of a local newspaper article. The article explained the purpose of the investigation and offered volunteers a reward of f 75,- in addition to the refund of electricity costs.
Occupants of over 50 homes volunteered; from these 50 homes, 20 were selected comprising a variety of typical Dutch homes. The selection was based on building structure and age of the home, and on the presence of (unvented) gas appliances.

Field operation

The equipment of the real-time monitoring program was operated by a field team of two people. At an appointed time (the occupants were briefed about the scope of the measurement during the home selection procedure) the equipment was installed in an empty room or attic in the home. From this room, the sampling lines were run to the kitchen, the living room, one bedroom and outdoors. The indoor sampling points were placed at breathing height. In the kitchen, the samples were drawn from three different sampling points. A metal cross was installed over the gas cooker in such a way

that the branches of the cross were in the flames of the burners. One of the thermo couples was connected to this cross. When a geiser was present, the second thermo couple was positioned directly above the appliance, in the flue gases.

After a warming-up period of at least 12 hours, the instruments were calibrated and the measurements were started. The monitors were checked and calibrated at least every second or third day. Visual inspection of strip chart recorders and of the printed read-outs of the micro computer of hourly average values allowed a direct check on the proper functioning of the equipment.
After several days up to two weeks, the equipment was dismantled and either installed in another home or brought back to the laboratory.

Data handling

Data cassettes of the micro computer were transferred to a mainframe computer and stored on tape. In general, a data set on a cassette comprised 2 or 3 days. When the interval between successive data cassettes from the same home was less than one or two hours and when the gas appliances were not used during this interval, the data sets from the cassettes were merged to form one file. Occasional erroneous data characters were skipped after inspection.

All data of each home were plotted for visual inspection of the material. Fortran programs were used for data reduction and analysis, after allowances for zero and span drift were made.

3.4. Week-long measurement program; material and methods

Instrumentation

The sampling devices for large scale week-long integrated measurements in occupied homes should be small, quiet, cheap, able to run unattended over the week and should require little maintenance. For NO_2, RSP and VOC such methods were more or less readily available from research in occupational settings. For CO and the SF_6 tracer experiments a combined sampling device was developed.

NO_2

For the NO_2 measurements passive diffusion tube samplers developed by Palmes et al. (1976) were used. These are small, 8 cm long, 1 cm inner diameter acrylic tubes with a stainless steel wire mesh, coated with the NO_2 absorbent triethanolamine, inserted at the closed end of the tube. Atmospheric NO_2 is transferred from the open end of the tube to the absorbent at the closed end according to Ficks first law, due to molecular diffusion.

After sampling, the absorbed NO_2 is analyzed spectrophotometrically at 540 nm, 30 minutes after adding 2.1 ml Saltzman reagent to the tube. Then NO_2 concentrations can be calculated with Ficks first law from the amount of collected NO_2, the dimensions of the tube, the sampling time and the diffusion coefficient (D; cm^2/sec) of NO_2 in air:

$$Q = - D.C.\frac{A}{Z}.t$$

where Q = quantity of transferred NO_2 (moles)
D = diffusion coefficient of NO_2 in air (cm^2/sec)
C = NO_2 concentration at the open end of the tube ($moles/cm^3$)
A = cross-sectional area of the tube (cm^2)
Z = the length of the tube (cm)
t = time of exposure of the tube (sec)

The diffusion tubes are used in numerous indoor air pollution studies, although the only field validation study has been carried out in 1 location in 3 homes (Apling et al. 1979). Tests of the U.S. National Bureau of Standards (Cadoff et al. 1979) and Warren Spring Laboratory in the U.K. (Apling et al. 1979) indicated a lower detection limit for the tubes of about 600 $\mu g/m^3$ over a sampling period of 1 hour (or 4 $\mu g/m^3$ over 1 week). Accuracy was demonstrated to be better than ± 10 %; precision was less than 4 $\mu g/m^3$ over one week sampling period.

Before and during the use of the tubes in the week-long integrated measurement program, extensive laboratory tests with the tubes were carried out. These tests were a corroboration of the precision of the diffusion tubes, but the calculated experimental diffusion coefficient of 0.115 cm^2/sec was consistently lower than the theoritical value of 0.154 cm^2/sec which is commonly used. In appendix B these discrepancies are discussed in more detail. The NO_2 concentrations in this study were calculated with the more conservative value of 0.154 cm^2/sec.

RSP

In 1981 an Ad Hoc Working Group of the International Standards Organization reported recommendations on size definitions for particle sampling, defining the alveolar fraction of particles i.e. the fraction which penetrates the non-ciliated lung airways (ISO 1981). This definition of the alveolar fraction is met by the American Conference of Governmental Industrial Hygienists (ACGIH) convention for the 'respirable fraction' of particles. According to the ACGIH convention samplers should collect 50 % of the particles with an aerodynamic diameter of 3.5 µm and 0 % of particles larger than 10 µm.
A convenient and compact instrument which follows the selection curve of ACGIH is the 10 mm cyclone presampler, combined with a filter sampler as second stage. At a sampling rate of 1.9 liter per minute, the presampler removes the non-respirable fraction of the particles.

The sampling units used in this study consisted of a Casella 10 mm cyclone, combined with a cassette containing a 25 mm diameter glass fibre filter (Whatmann ME 44, pore size 5 µm). Air was sampled over the filter at a flow of 1.9 l/min with a Dupont P 2000 personal sampling pump connected to the mains with an adapter. For noise reduction the pumps were placed in a wooden box, lined with isolation material. Before and after sampling, the filters were humidity conditioned and weighed at 45 % relative humidity. Series of duplicate samples (n=25) demonstrated a precision for the method, expressed as coefficient of variation, of 5-8 %.

To get an impression of the magnitude of peak-concentrations of RSP, a TSI Piezobalance, for instantaneous respirable mass measurements of particles, was used.

VOC

VOC were sampled on SKC Inc. sampling tubes packed with 100 mg activated charcoal as absorbent with a back up section of 50 mg charcoal. Samples were collected at a flow of ± 100 ml/min. with Dupont P 200 and P 4000 personal sampling pumps attached to the mains. Pumps were placed in wooden boxes for noise reduction.
At the National Institute of Public Health and Environmental Hygiene in Bilthoven the collected samples were desorbed by counter-current elution with 1 ml CS_2 and analyzed by high resolution capillary gaschromatography with flame ionization detection (Packard 433; column:50 m, 0.32 mm ID., fused silica, coating 0.4 µm CPsil5; carrier gas:He, flow 1.2 ml/min; temp.

program. 0-180 °C, 2 °C/min; detector FID, det.temp. 250 °C). For identity confirmation, several samples were also analyzed by capillary gaschromatography/quadrupole mass spectometry.
With this sampling and analytical method, 45 of the most abundant coelutent free VOC in the boiling point range of 70-270 °C could be determined quantitatively on a routine basis. The detection limits of the method were between 0.4-4 $\mu g/m^3$ for the chlorinated hydrocarbons and about 0.3 $\mu g/m^3$ for the other compounds. Duplicate field samples (n=10) showed a coefficients of variation for the different compounds of 15-20 % for the method.

CO and SF_6

For the CO and SF_6 measurements a device was developed which sampled air in 10 liter aluminium-polyethylene foil bags (Linde Plastigas). The sampling bag was connected with a 1/16 inch stainless steel tube to a Charles Austen model L12 pump with a 1/16 inch polyethylene inlet line. The pump was switched on and off by a timer (S-system electromatic, Model SC 185-220) to run for one out of every ten minutes over the week-long sampling period. A Hoke 1335 G 4 B metering valve was used to adjust the sample flow at about 10 ml/min.
Laboratory experiments showed virtually no loss of CO and SF_6 after storing the sampling bags for 7 to 8 days; consequently no desorption of CO and SF_6 was observed after evacuation of the sample and flushing of the bag with zero air.
The bags content was analysed gaschromatographically on CO (after catalytic reduction to CH_4) with flame ionization detection and on SF_6 with electron capture detection. (CO: Packard 5710 A; column: 1m,1/8 inch, Molsieve 5A, 40/60 mesh, temp. 110 °C; carrier gas: H_2, 30 ml/min; cat.: Ni, 20 cm, 1/8 inch(Chrompack), temp. 300 °C; detector FID, det. temp. 250 °C.// SF_6: Packard 5710 A; column 1.8 m, 1/4 inch, Molsieve 5A, 40/60 mesh, temp. 70 °C; carrier gas: N_2, 46 ml/min; detector ECD, det. temp. 200 °C). Detection limits for CO and SF_6 were 1 mg/m^3 and 0.1 $\mu g/m^3$ respectively. Initial tests at the laboratory indicated a coefficient of variation for duplicate samples of about 10 %, but 15 duplicate field samples had a coefficient of variation of about 15 %.

The SF_6 emission (in the kitchen) was similar to that in the real-time monitoring program. Instead of pure SF_6, the cylinders contained air with 15,000 mg/m^3 SF_6, because compared to the infrared analyzer, the gaschromatographic method has a lower detection limit.

Additional information

For descriptive purposes and for the development of a predictive model for indoor pollutant levels, additional information was needed about the factors which control indoor air quality. These factors can be measured at different levels: in its simplest form a measure for such factors is the presence or absence of indoor pollutant sources or of ventilation provisions.

A more sophisticated level of measurement of indoor pollutant production (similar arguments apply to ventilation) can be achieved by incorporating not only the presence of a source but also its actual use, for instance the frequency (and duration) of gas cooking or smoking.
Extensive inspection lists, questionnaires and diaries were developed to determine indoor pollutant production and elimination, at different levels of sophistication. The additional information was gathered at different levels in order to obtain an optimal predictive model (in terms of accuracy of the prediction) for indoor pollutant levels, with as simple a form of information as possible.

The inspection list contained items about the home:
- type of home
- number of floors
- number of rooms
- volume of kitchen, living room and one bedroom
- type of kitchen (open or separated from living room)
- presence of vented and unvented gasappliances
- presence of doors, windows and ventilation provisions in kitchen, living room and bedroom.

The questionnaire consisted of questions on occupants and their activities:
- family size
- socio-economic status
- use and maintenance of the geiser
- frequency and duration of use of gas cooker and oven
- frequency and duration of use of ventilation provisions and of interior doors between kitchen, living room and bedroom
- complaints about draught and moisture
- application of insulation and crack sealing materials
- smoking habits of family members
- frequency of vacuum cleaning
- use of solvents and several other consumer products.

The diaries were designed to assess the actual occupant activities during the measurement period:
- number of persons in the home*
- use of unvented gas appliances*
- use of ventilation provisions and interior doors*
- number of hot meals which were prepared**
- number of cigarettes which were smoked**
- use of the vacuum cleaner**

* on hourly basis
** on half a days basis

Selection of homes

Given the available manpower and instrumentation it was considered that 200-300 homes could be handled in two winters, when in each winter the survey would be restricted to one town.
First, a town was selected for the first winter in the vicinity of Wageningen (from where the measurement program was run) to gain experience with the field work. The town of Ede, with about 50,000 inhabitants, was suitable for this purpose. The older homes in this town, built before the Second World War, however, were not very typical for the Netherlands. Therefore, only post-war homes were selected in Ede.
Second, it was assumed that older homes built before World War II, especially in the inner-city of large towns, might deviate unfavourably from newer homes with respect to indoor air quality, due to a different geometrical design, smaller volume, crowding and a more frequent presence of unvented gas-fired water heaters. To incorporate these types of pre-war homes, five neighbourhoods in the inner-city of Rotterdam were selected as study area for the second winter.

In the second winter, an additional group of homes was selected in Ede in neighbourhoods built after 1976. In this group of homes only volatile organics were determined; the objective was to assess whether new building materials would lead to higher volatile organic levels than those found in older homes.
Both in Ede and in Rotterdam homes were randomly selected from the home register of the towns. To avoid non-typical types of homes, only homes with three to six rooms were selected.
The homes selected in this way cannot be considered as a representative sample of the total housing stock of the Netherlands; they do, however, represent the situation that exists in large numbers of typical Dutch homes.

Approach of the occupants

Between October and April, each week 10 to 20 introductory letters were sent to homes from the selected sample. The letter introduced the investigation and its purpose to the occupants, and announced the visit of a team of two field workers. After this letter, the homes were visited, a further explanation on the scope of the measurements was given and the occupants were invited to participate. If the occupants volunteered to cooperate, an appointment was made to install the equipment.

When occupants were not contacted after three home visits, attempts were made to contact the occupants by phone, if a door-plate with the occupants name was available and the occupants were listed in the telephone book.If all this failed, the occupants were listed as non-responders.

Installing and collecting the equipment

In each week one or two days were reserved to install the equipment in 5 or 10 homes; one day was reserved to collect the equipment. This meant that for logistic reasons the week-long measurement period covered 5 to 7 days including the weekend.

One of the two field workers operated the sampling devices and determined the flow rates with flow rators and soap bubble meters. After checking the proper functioning of the samplers, they were installed in the different locations at breathing height away from obvious pollutant sources and ventilation provisions.
NO_2 samplers, available in large quantities, were used in kitchen, living room and bedroom. RSP and VOC samplers were used in the living room only. The CO/SF_6 samplers were used in the kitchen and living room. Instantaneous RSP measurements were carried out with the TSI Piezobalance in the kitchen, living room and bedroom. A sketch of the sampling locations (kitchen, living room and one bedroom) was made, including the position of the sampling points, gas appliances, fire places, doors and ventilation provisions; then the inspection list was filled in.

The second field worker administered the questionnaire; the occupant who did most of the housekeeping was asked to respond. After the questionnaire, the same occupant received the diary forms and a verbal and written explanation was given about how to fill in the diaries. Finally, an appointment was made to collect the equipment 5 to 7 days later. On average, the installation visit lasted about one hour.

When collecting the equipment, one field worker delocated the equipment and determined the flow rates of the samplers, while the other administered a short questionnaire on the use of possible sources of VOC. The diary forms were checked on obvious indistinct or incorrect entries.
At both visits notes were made about the position of windows, ventilation provisions and (interior) doors during the visit.

Data handling

Samples of the inorganic pollutants were analyzed at the laboratory in Wageningen, without information about the sampling period or the location. The results of the NO_2 assay and the weight collected on the filters were later combined with data on sampling time and volume, to give concentration values. During the field work it appeared that (in contrast to the initial tests) the CO/SF_6 sampling units did not collect as much sample as would be expected on the basis of the begin and end flow rates. The volume of the sample in the bags was therefore rated from 1 to 4. Bags with a score of one were discarded as having insufficient sample. Differences of more than 20 % between begin and end flow rates of samplers and SF_6 emission were considered unacceptable.

The diary information on the use of gas appliances etc. were aggregated over the measurement period and standardized on a one week (168 hour) basis. The diaries were rated on a 3 point scale based on inconsistencies, unclearities or insufficient detail. Diaries with a score of one (insufficient) were discarded.

All the data, including questionnaires and inspection lists were coded by hand, card punched, transferred to a DEC 10 computer and stored in 1022 Data Management Systems-sets and system files of the SPSS and BMDP statistical software packages.

The VOC samples were sent to the National Institute of Public Health and Environmental Hygiene for analysis. The screened concentration data were received back on computer tape and merged with the other data sets.

Data screening

Extensive data screening procedures were carried out on the data sets to check on abnormalities and erroneous data entries. Among others, these

procedures consisted of checks on minimum and maximum values of nominal or ordinal scale variables (the dichotomous variable with code 1 or 2 cannot take the value of 3), checks on outliers of interval and ratio scale variables and numerous cross-checks (when the family size is 2, the maximum number of smokers in the household is 2; when there is no exterior door in the kitchen, the diary variable 'use of exterior kitchen door' cannot take a positive value).

Samples of the diaries were tested on face validity by comparing the use of ventilation provisions as noted during the home visits by the field teams with the frequency and time of use of these provisions according to the diaries. This crude test revealed no clear irregularities.

All unexpected values detected in the screening procedures and subsequent data analysis were checked in the original data forms; depending on the results of the checks, values were left unchanged, corrected or discarded.

3.5. Repeated measurement program; material and methods

Instrumentation

The repeated measurement program consisted of week-long integrated measurements of NO_2 in kitchen, living room and bedroom in 15 homes every other week over a period of one year.
In addition, week-long RSP and VOC samples were taken every other week during half a year, in the living room in a subgroup of four homes. The instrumentation for the repeated measurements was the same as for the week-long integrated measurement program. CO and SF_6 measurements were excluded from this program because the sampling devices were more intrusive than the samplers for the other pollutants. RSP and VOC measurements were restricted by the limited availability of sampling devices.

A special group of repeated measurements of VOC levels in newly built homes, before and during occupancy, had the objective to distinguish between the impact of building materials and that of occupant activities on VOC levels.
The VOC measurements in the unoccupied newly built homes were carried out over 8-hour periods with battery powered Dupont P 4000 pumps because electricity was not available. The sampling rate was 1 l/min. In the occupied homes week-long measurements with the common sampling arrangements were carried out.

Home selection

Employees of the Department of Air Pollution and the Department of Environmental and Tropical Health were asked to participate in the repeated measurement program. From the 15 volunteers who participated in the NO_2 repeated measurements, 2 non-smokers and 2 smokers were invited to participate in RSP and VOC measurements.

For the measurements in newly built homes cooperation was obtained from the "Dutch Municipalities Building Fund", who provided access to 11 homes of a building project of government subsidized private homes and mediated with the future occupants. The 11 homes were all identical and were constructed in the same building stream.

Field operations and data handling

At the start of the repeated measurement program the volunteers received a briefing and demonstration about the operation of the sampling devices. For RSP and VOC samples the occupants were supplied with flow rators and soap bubble meters.
Every other week the volunteers received fresh sampling units, which they installed at home. After a sampling period of one week the samplers were brought back to the laboratory.

In the newly built homes repeated measurements were carried out at 6 phases:
- in empty homes, just before occupation;
- just after floor- and wall covering was carried out;
- just after the homes were furnished;
- in occupied homes, during the first week of occupation, after 1 month and after 3 months of occupation

The samples and data of the repeated measurement program received a similar treatment as those of the week-long integrated measurement program.

3.6. Statistical analysis

The statistical analysis of the data was carried out with SPSS (Nie et al. 1975, Hull and Nie 1981) and BMDP (Dixon et al. 1981) routines. Apart from several descriptive and screening statistics, the major statistical techniques which have been used were multiple regression analysis, analysis of variance and factor analysis.

Multiple regression analysis

The multiple regression analysis was mainly used for the development of models to explain and predict the indoor pollutant levels measured in the week-long integrated measurement program (the dependent variables) with the characteristics of the home and occupants (the independent variables).
The relationship between a dependent variable y and n independent explanatory variables x_1, x_2, ..., x_n in a linear model is expressed by formula:

$$\underline{Y} = \beta_0 + \beta_1 + \beta_2 + ... + \beta_n + \varepsilon$$

where $\underline{Y}$ is the dependent or response variable and ε the error term. The regression coefficients $\beta_i (i=0,1,...,n)$ can be estimated with the method of the least squares, by $b_0,...,b_n$, so that the expected value of the dependent variable ($\hat{Y}$), given the values of the independent variables can be expressed by the following regression equation:

$$\hat{Y} = b_0 + b_1x_1 + b_2x_2 + ... + b_nx_n$$

where b_0 is the constant or intercept of the equation; b_1, b_2, ..., b_n express the marginal effect of x_i on the dependent variable, i.e. the increment in $\hat{Y}$ which corresponds with an unit increase in the independent variable x_i when all other variables are held constant. The regression coefficients can also be expressed as standardized regression coefficients by standardization of the dependent and independent variables to unit variance (i.e. standard deviation of 1). These standardized regression coefficients make it possible to compare the relative effect on the dependent variable of independent variables which have been measured on different units, for instance kitchen volume in m^3 and tobacco consumption in cigarettes per day.

To obtain an optimal regression equation (in terms of explained variance) with as few independent variables as possible, subsets of all independent variables were selected in the data analysis, using the 'stepwise' selection procedure of the SPSS 'New Regression' routine. In many cases, other selection procedures, like 'backward elimination' were also used, generally yielding the same results as the 'stepwise' procedure. In this thesis, only the results of the 'stepwise' procedure will be presented.

A summary statistic for the adequacy of the fit of the regression model to the data is the multiple correlation coefficient R. When squared, R^2 expresses the proportion of variance in the dependent variable - the pollutant concentration for instance - that can be accounted for, or be explained by the independent variables in the model. The R^2 statistics presented in this study are all adjusted for the number of independent variables in the regression equation and the number of cases.

Another important statistic for the evaluation of the regression equation is the standard error of estimate, SEE. The SEE is the square root of the error variance. It can be interpreted as an average error in predicting the dependent variable from the regression model.
In other words, the SEE is a measure for the accuracy of the prediction expressed in the absolute units of the dependent variable.
Without a predictive model, the best prediction of the value of y, for instance the NO_2 concentration in a home, would be the mean of y over all homes (assuming a normal distribution of y over the homes). The standard deviation σ_y in the group of homes describes the accuracy of this prediction. The gain in accuracy obtained by using a predictive (regression) model instead of the mean of y, can be established by comparing the SEE of the predictive model to σ_y. A measure for the gain in accuracy is the coefficient of alienation (CA), which is the ratio of the SEE to σ_y (Rozenboom 1966). The CA is related to the multiple correlation coefficient R, but the CA is a more direct measure for the gain in accuracy than the R^2.
Due to logarithmic transformation of the dependent variable, the interpretation of the SEE is not as straightforward as described above. Therefore the CA instead of the SEE will be presented for the regression models in this report, in addition to the R^2.
(For backgrounds on multiple regression analysis see for instance Chatterjee and Price (1977) and Snedecor and Cochran (1980)).

Analysis of variance

Analysis of variance was used to estimate the variability in indoor pollutant levels in time and place from a number of repeated measurements in homes.
The observed variance in pollutant levels (σ_0^2) from repeated measurements can be decomposed in error variance (σ_e^2), the within-home variance in levels, and in true variance (σ_t^2), the variance in levels between different homes (Snedecor and Cochran 1980):

$$\sigma_0^2 = \sigma_t^2 + \sigma_e^2$$

The reliability coefficient (RC) expresses the ratio of true variance to the observed variance:

$$RC = \frac{\sigma_t^2}{\sigma_0^2}$$

The reliability coefficient will be high when the differences between repeated measurements within homes are small as compared to the differences in (average) levels between different homes. In this case a single measurement will give a good impression of the typical level for a particular home; it makes it possible to discriminate between indoor levels in homes with little effort. A low reliability coefficient indicates that it is difficult to establish differences in indoor pollutant levels between homes with only one or a few measurements; several repetitions of the measurements will be required to discriminate between pollutant levels of different homes.
The RC can be calculated for a single measurement and for the mean of several repeated measurements, the latter always being higher. In the case of only 2 repetitions of the measurements (test-retest situation) the RC equals the correlation coefficient between the first and second measurement (Snedecor and Cochran 1980).

Factor analyis

Factor analysis was used to delineate possible patterns in the VOC concentrations in the homes of the week-long integrated measurement program. The rationale behind the factor analysis in this case is that, when several compounds share the same source, the use of this source will elevate the levels of all these compounds; thus, when the source is powerful enough these VOC levels will be highly correlated. The objective of the factor analysis is to identify factors by clustering the highly correlated compounds together in one or a few groups. Preferably, each factor should contain compounds which are highly correlated with each other but not with other compounds and each compound should be involved in as few different factors as possible. The factors therefore hypothetically describe VOC which are produced by the same source or group of sources and may provide clues about the nature of the sources. The degree of involvement of a variable in a factor is expressed by the factor loadings, which can take values between 0 (no involvement) and !1 (highly involved in the factor). (For backgrounds on factor analysis see for instance Rummel (1967) and Frane and Hill (1976)).

Distribution of the data

The statistical techniques which were used are based on the assumption of a (multivariate) normal distribution of the data. This assumption is seldom met in survey type indoor air pollution studies. More often a lognormal

distribution of pollutant levels is observed, which calls for a logarithmic transformation of the data to better satisfy the assumption of normality.

The logarithmic transformation has the disadvantage that the results, especially regression coefficients, are more difficult to interpret; in addition, the effect of independent variables on the dependent variable in regression analysis is no longer additive but multiplicative. The transformation may also mask peculiarities in the data that may be of special interest.

Although the statistical tests which were used are considered robust, the violation of the assumption of normality may result in unstable parameters dominated by a few extreme values; furthermore it may produce flattered estimates of statistical significance and of the amount of explained variance.

In the process of statistical analysis most calculations were carried out on the crude data as well as on logarithmically transformed data; sometimes rank-order scale data have been used. To avoid an abundance of information, only a selection of the calculations will be presented. In general, the results of calculations after a log transformation of the data will be presented when the distribution of the data and residual statistics indicated that a log transformation would be appropriate.

Significance levels

Ideally, the significance level of all parameters of all calculations should be presented so that the reader can make his own judgement. Again this would lead to an abundance of information. Instead, a significance level of $p \leq 0.05$ has been adopted as a formal criterium. Thus, a stricter criterium was used in calculations over a small number of cases (because of many missing values) than in calculations with a large number of cases. Furthermore it should be noted that differences which are statistically significant may not always be substantial.
The exploratory character of the study (i.e. the lack of isolated well-defined a priori hypotheses), the selection of the presented statistical calculations and the arbitrary choices about the appropriateness of data transformation which had to be made, have consequences for interpretation of the significance level of the presented calculations. It should not be interpreted too literally as "the probability that a given statistical relationship would occur by chance alone". The significance level of 0.05 is used more as a convenient fixed criterium to select information, which

may be too strict or too lenient in some cases. (For a discussion on the significance of tests of significance see Morrison and Henkel (eds.):" The Significance Test Controversy" 1970).

PART 2.

CHAPTER 1. INTRODUCTION

Part 2 of this report contains the results of the 3 main measurement programs which were described in part 1, chapter 3.
Chapter 2 of part 2 concerns general characteristics of the homes and its occupants, and indoor pollution production and ventilation of the homes.
For the real-time monitoring program, the general characteristics of the homes and its occupants are meant as a brief description of the homes which were studied. Part of the interpretation of the results of the real-time measurements were based on qualitative examination of data plots. Since plots of the measurements in 1 home over a period of 24 hours already covers 5 pages, only examples of these plots can be presented.
The characteristics of the homes and occupants of the week-long measurement program are presented as a description of the "study population" and as an illustration of the differences between the age-groups of homes. The presentation of data about indoor pollution production and about ventilation of the homes are restricted to those features which are used for, and may contribute to, the explanation and prediction of the measured pollutant levels in the homes. Thus the data about use of ventilation provisions and gas appliances are presented only for the post-war homes in Ede and the pre-war homes in Rotterdam, while for the use of possible indoor sources of VOC, data will also be presented for the less than 6 years old homes in Ede.

The results of the CO, NO_2, RSP and VOC measurements will be given respectively in the chapters 3 to 6. In each chapter the results of the 3 main measurement programs will be treated separately. When appropriate, results of related studies will be presented as well, as was mentioned in part 1, section 3.1.

CHAPTER 2. GENERAL CHARACTERISTICS OF THE HOMES UNDER STUDY

2.1. The real-time monitoring program

Description of the homes

In 12 of the 20 originally selected homes succesful measurements were carried out over periods of 135 to 273 hours. The measurements in the other homes were cancelled or discarded because of malfunctioning of the equipment, illness of occupants or complaints about the noise of the equipment. The measurement period and some characteristics of the 12 homes and its occupants are presented in table 6.

All the homes were located in a suburban area, in or near Wageningen. The single family homes were semidetached or terraced homes. In these homes, the bedroom from which air was sampled, was located on the second floor. Kitchen and living room were located on the first floor in all the homes. Casement windows and/or vent lights were present in all rooms.

Use of unvented gas appliances

The primary purpose of the data on the use of the unvented gas appliances was to examine the influence of appliance use on the indoor CO and NO_x levels. The temperature above the gas appliances, monitored with the thermo couples, were plotted versus time of day and analysed qualitatively in combination with plots of the pollutant levels in the three rooms.
The plots showed that in each home, the frequency and duration of appliances use was more or less consistent from day to day. When different homes were compared, more differences than similarities were observed in the patterns of use.
In general, the gas cooker was used 2 to 6 times a day with a regular day to day pattern. Characteristic of the geiser was its frequent and irregular use of 10 to 35 times a day for periods of about 1 or 2 minutes. Figure 2 shows an example of the pattern of use over 1 day of the appliances in home 2 and 3.
In home 3, the pilot burner of the geiser was used only during the day; in the example in figure 2, the pilot burner was lighted at about 7.30 a.m. and shut off at 7 p.m.
In the other homes equipped with a geiser, the pilot burner was used day and night. The pilot burner of the cooking range was used in none of the homes.

In home 7, the oven was used to heat up the living room (figure 3). The heated air from the oven was directed to the living area with a fan; when the desired temperature was reached in the living room, the central heating installation was used to maintain this temperature.

Ventilation of the kitchen

In 10 of the homes, succesful measurements of the SF_6 concentration in the kitchen were obtained over periods of several days. For these homes, the SF_6 concentration in the kitchen was plotted versus time of day; kitchen air change rates (ACR) were calculated from the SF_6 emission rate, the 30-minutes average SF_6 concentration and the volume of the kitchen, as described in appendix A. THe ACR expresses the number of times per hour the air in the room is completely replaced by (fresh) air. The TI expresses the efficiency of a ventilation system in protecting a given position in that system from exposure to airborne contaminants which are released at another position in the ventilation system. Under conditions of complete mixing, the TI is the reciprocal of the product of the ACR and the volume of the room. Thus, the higher the ventilation and the larger the volume, the lower the TI will be (cf. appendix A). These ventilation rates were plotted versus time of day in combination with the SF_6 concentrations.
In several cases, these plots revealed cyclic patterns and rapid changes in the SF_6 concentrations. The rapid changes in SF_6 concentration often coincided with the use of gas appliances (figure 4). Note that the pattern of the ventilation rate versus time is smoother than that of the SF_6 concentration because the ventilatory rate is calculated from 30-minute average SF_6 concentrations.
From interviews with occupants it appeared that the rapid changes in SF_6 concentrations also occurred at times when the use of ventilation provisions was not changed. This indicates that the changes in SF_6 concentration reflect mixing of stratified kitchen air by human activity and by the use of gas appliances, as well as changes in ventilation of the kitchen. The cyclic pattern of the SF_6 concentration, which was observed in several homes especially during the night, was probably the result of convection of the kitchen air caused by the heat of the refrigerator motor.
The operation of the refrigerator motor was monitored in several homes by placing an additional thermo couple on the shield of the refrigerator motor. It appeared that the cyclic fluctuations of the SF_6 concentrations had the same frequency as the repeated operation of the refrigerator motor (figure 5).

To smoothen the effect of short-term fluctuations in SF_6 concentration on the ACR, 4-hour average ACR and transfer indices (TI) were calculated from 4-hour average SF_6 concentrations. Table 7 presents the overall mean and range of the ACR and the overall mean TI of the measurement period in the 10 homes.
Large differences in the 4-hour average ACR were observed both within homes and between different homes. The highest 4-hour average ACR in a kitchen was about 2-6 times the lowest ACR in that kitchen. The homes with an overall mean ACR, smaller than 1 h^{-1} all had an open kitchen and therefore a large kitchen volume. When judged on the TI from a hygienic point of view, home 10 had the best (= lowest) value. This home had the highest capacity to dilute and disperse pollutants generated in the kitchen. Judged on the ACR however, home 10 is only the sixth best in rank.

2.2. The week-long measurement program

2.2.1. Response

In the first winter, introduction letters were sent to 326 post-war homes in Ede. In the second winter 417 inner city pre-war homes in Rotterdam and 149 less than 6 years old homes in Ede were approached. The response rates for the 3 age-groups of homes are given in table 8.
More than 50 % of the occupants which could be contacted, agreed to cooperate in the study. In Rotterdam the positive response was deflated by city renovation and renewal activities; 7 % of the homes to which an introduction letter had been mailed were empty or demolished.
In addition several occupants in Rotterdam refused to participate because they would leave their home in the near future due to renovation of the home. In 19 of the homes in Rotterdam the occupants spoke the Dutch language unsufficiently to participate in the study.

Because the homes were visited during office hours, households with members who were at home during these hours had the highest chance to be contacted. Households with full-time working members will therefore be underrepresented in the study population.

The distribution of several characteristics of the post-war homes in Ede in the study population could be compared with the official data from the municipality of Ede (table 9). Table 9 shows that unmarried occupants were underrepresented in the study population, while married and widowed occupants were overrepresented. Private homes and single family homes were overrepresented in the study population by 4 %. These differences between

study population and the official data for the study area were probably the result of the same selection mechanisms.
Married occupants - especially those who have young children - and elderly (widowed) people had a higher chance to be contacted during office hours than other groups of occupants. The private homes in Ede are often of the single family type of home. Because these homes are often occupied by married occupants, those type of homes are overrepresented. Furthermore, it appeared during the fieldwork that, in general, people who owned their home were more interested to participate than people who rented their home.

2.2.2. General description of the study population

In table 10 some general characteristics of the occupants and their homes are presented as a summarized description of the study population. The table illustrates some of the many differences between the 3 age groups of homes, which are the result of (cultural) differences between the suburban (Ede) and urban (Rotterdam) study areas and of the developments in the housing stock.
Several of the general characteristics were interrelated. For instance, as mentioned before, private homes are often of the single family type and occupied by married people. In general, these homes are large and more often equipped with a range hood, mechanical ventilation system and (when a geiser is present) a geiser vent than other homes.

2.2.3. Use of ventilation provisions in post- and pre-war homes

In the discussion about ventilation provisions, the term ventilation appliance is used to describe mechanical operated appliances such as the window fan and range hood. The term ventilation provision is used for windows and exterior doors which can be opened by the occupants.
Three types of ventilation provisions were identified in the study: vent lights, casement windows and outside doors.
Ventilation grills were treated as vent lights; sash windows were treated as casement windows in the data analysis.

In Ede, 8 homes had no ventilation provisions or appliances in the kitchen. These homes were part of a flat for elderly people and had a small electric cooking facility. The living rooms and bedrooms in Ede all had one or more ventilation provisions.
In Rotterdam, 1 kitchen, 5 living rooms and 9 bedrooms had no ventilation provisions. All these rooms had an open connection to other rooms which did

have one or more ventilation provisions.

Table 11 presents the percentage of homes in Ede and Rotterdam in which none of the ventilation provisions in kitchen, living room and bedroom were actively used for ventilation during the heating season, according to the answers of the questionnaires.
The kitchen window fan was only used regularly in about 50 % of the homes in which this appliance was present. The range hood, when present, was used regularly in 80-90 % of the homes.
Two reasons for not using ventilation provisions in a room were often heard during the field work, especially in Rotterdam. The first reason was that the room had an open connection with other parts of the home where the occupants did use ventilation provisions. The second reason was draught. In Ede 29 % of the respondents often experienced draught, against 44 % of the respondents in Rotterdam.

The use of ventilation provisions during the measurement week were analysed as follows. For each ventilation provision in a room, the duration of use was aggregated over the measurement period, standardized for a period of 168 hours (1 week) and expressed as the percentage of time in which the ventilation provision had been used. Then, the variable 'total use of ventilation provisions' was formed for each room, by summation of the use of different ventilation provisions which were present in the room. For instance, when the vent light in the kitchen was used 30 % of the time and the kitchens casement window was used 5 % of the time, the variable "total use of ventilation provisions in the kitchen" would take a value of 35 %.
No allowances could be made for the time of day at which ventilation provisions were used or whether windows were set ajar or were wide open. In some homes, the sum of the use of vent light and casement window was more than 100 %, which means that these ventilation provisions were open at the same time during a part of the measurement period.
Figure 6 presents the frequency distribution of the total use of ventilations in the 3 rooms in Ede and Rotterdam. This figure shows a more or less bimodal distribution of the use of ventilation provisions, which is most pronounced in Rotterdam in the kitchen and bedroom. This indicates that 1 group of occupants only used the ventilation provisions during relatively short periods of time, while another group used the ventilation provisions (almost) the entire measurement period. In Ede, the occupants used the ventilation provisions during longer time periods than those in Rotterdam, especially in the bedroom.
The diary results were in agreement with the questionnaire information, from which two kinds of ventilation behaviour could be identified; one group of respondents practiced airing of the rooms during relatively short

periods of time, while others maintained a more permanent ventilation by a more or less continuous use of ventilation provisions.
The geometric mean and range of the average daily use of range hood and kitchen window fan during the measurement period according to the diaries, in the homes equipped with these appliances, is given in table 12. On average, the range hood was used twice as long in Ede as in Rotterdam. This is in agreement with the questionnaire information. The window fan was not used in 50 % of the homes in Ede and in 58 % of the homes in Rotterdam, which of course affected the geometric mean of the use of these appliances.
The time during which interior doors of kitchen, living room and bedroom were open was also assessed with the diaries. Respondents were asked to record the periods when doors were open during more than 5 minutes. Thus, the time necessary to pass a door was not recorded.
The use of interior doors was calculated in a similar way as for the use of ventilation provisions and expressed as the percentage of time during which the doors were open. When occupants had removed the interior door (which was done quite often) this was considered in the data analysis as if the door was open permanently.
The geometric mean of use of interior doors in the 3 locations in Ede and Rotterdam is given in table 13. The range in the use of interior doors was from 0 % to 100 % in all the locations in both towns.
Especially interior doors of the kitchen were open during a considerable fraction of the time; the geometric mean was 29 % of the time in both towns. The interior doors of the other locations were, on average, open during a few percent of the time. In Rotterdam, the interior doors of the bedroom were on average open twice as long as in Ede. This is mainly because in Rotterdam the bedroom door was removed in 10 % of the homes, while in Ede this was only the case in 1 % of the homes.

2.2.4. Use of gas cooker and geiser in post- and pre-war homes

The use of the gas-fired cooking range and geiser was determined with the questionnaires and diaries. The questionnaire information, however, had only limited use to assess differences in the use of the gas appliances. The respondents felt it difficult to report a typical duration of use.
In about 80-90 % of the homes hot meals were prepared virtually daily; the preparation time for hot meals took between 30 to 60 minutes in about 75 % of the homes.

In the diaries, the use of gas appliances was only registered if the appliances were located in living areas of the home. Appliances (mostly geisers) in attics, cellars or sculleries which were isolated from living

areas were not considered, because it is unlikely that these appliances would affect the indoor pollutant levels.
For geisers, only periods of prolonged use, for instance for the shower, were filled in in the diary forms.
The duration of use of gas appliances during the measurement period, according to the diaries was standardized for a period of 168 hours and expressed as the average daily use in minutes.
Table 14 presents the geometric mean and range of the use of gas appliances in Ede and Rotterdam.
A wide range in use of gas appliances is apparent in table 14. On average, the gas cooker was used about 1 hour a day.
The oven, when present, was used in only 50 % of the homes in Ede and in 37 % of the homes in Rotterdam during the measurement period.
Prolonged use of the geiser was on average 16 minutes per day in Ede and 6 minutes per day in Rotterdam. The difference in geiser use between the 2 towns is probably the result of differences in hot water supply for the shower.
In Ede 85 % of the geisers supplied hot water for the shower, while in Rotterdam only 67 % of the geisers was connected to the shower.

2.2.5. Tobacco consumption in post- and pre-war homes

Three measures were used to describe tobacco smoking in the homes: the number of smoking occupants, tobacco consumption according to the questionnaire and tobacco consumption according to the diaries. Tobacco consumption was calculated by summation of the number of cigarettes, cigars and pipes smoked per day inside the homes. For convenience, it is expressed as the number of cigarettes smoked per day.
In Ede 60 % of the homes had 1 or more smoking occupant. In Rotterdam this was 66 %. According to the diaries, the geometric mean tobacco consumption was 4 cigarettes per day (range 0-75) respectively 6 cigarettes per day (range 0-51) in Ede and Rotterdam.
Table 15 presents the results of the daily tobacco consumption according to the diaries in the 2 towns, broken down by the number of smoking occupants. The table shows that in non-smoker's homes on average 1 or 2 cigarettes per day were smoked during the measurement period, presumably by guests. This figure should be interpreted with some care because further analysis indicated that the tobacco consumption in homes of non-smokers was not spread regularly over the days of the week, but was concentrated on only 1 or 2 days in most cases.
The number of smokers in the home explained 60 % respectively 54 % of the variance in the logarithm of the tobacco consumption during the measurement

period in Ede and Rotterdam. A large overlap in tobacco consumption in successive groups of the number of smoking occupants was observed (cf. table 15).
The squared correlation coefficient (R^2) between the logarithm of the tobacco consumption according to the questionnaires and that of the diaries was about 0.80 in both towns, indicating a good agreement of the 2 measures for tobacco consumption.

2.2.6. Use of possible indoor sources of VOC in 3 age-groups of homes

The use of possible indoor sources of VOC during the measurement period was assessed by a questionnaire administered at the end of the measurement period. Questions were asked about the use of a wide array of (consumer) products in the home. The questionnaire had an exploratory character; examples of various kinds of products like solvents, paint, adhesives, aerosol sprays etc. were mentioned by the field workers and the responders were invited to report the use of any product or material they might think relevant.
For the data analysis, the products reported by the respondents were combined into groups. Table 16 presents the percentage of homes in which the product groups were used during the measurement period. Respondents were also asked to quantify the amount of products used, but a reliable quantification appeared not possible, among others because the product was used by another occupant than the respondent. By consequence, the classification of product use is crude. The use of solvent for instance, may apply to the cleaning of a paintbrush or to the cleaning of a large surface.

2.2.7. SF_6 tracer gas experiments in post- and pre-war homes

In Ede successful tracer gas experiments were carried out in 72 kitchens and 73 living rooms. In Rotterdam these measurements were successful in 69 kitchens and 73 living rooms.
From the results of the tracer gas experiments air change rates (ACR) and transfer indices (TI) were calculated as described in appendix A. Table 17 shows the geometric mean and range of the TI and ACR in the kitchen and the TI in the living rooms in the two towns; for homes with an open kitchen/living room the ACR of the living room was also calculated.
The geometric mean TI had comparable values in Ede and Rotterdam in the kitchen as well as in the living room. As could be expected the TI of the living room was lower than that of the kitchens.

High kitchen ACR were observed more frequently in Rotterdam than in Ede. Kitchen ACR of 15 (h^{-1}) were observed in 3 homes in Ede and 11 homes in Rotterdam. In all these homes the interior door of the kitchen was either removed or left open during the entire measurement period. The kitchen with the highest ACR of 24 (h^{-1}), observed in Ede had an TI of 0.5 (min/m^3), the same value as the geometric mean of all kitchens in Ede. The kitchen with the highest ACR in Rotterdam had a TI of 0.2 (min/m^3). In this kitchen the vent light as well as the interior door were open all the time during the measurements. These comparisons of the ACR and corresponding TI in the kitchen clearly illustrate the difficulties which occur in determining ACR in occupied homes, when the effective ventilation volume cannot be determined accurately and deviates from the physical volume of the room.

Multiple regression analysis with the logarithm of the TI in the living room as dependent variable and the logarithm of the TI in the kitchen, the number of doors between kitchen and living room and city as independent variables showed that the independent variables explained 38 % of the variance in the dependent variable (table 18). After adjustment for the logarithm of the TI in the kitchen and the number of doors between kitchen and living room the logarithm of the TI in the living room was significantly higher in Rotterdam than in Ede, indicating a higher exchange of air from kitchen to living room in Rotterdam.

2.3. Repeated measurement program

The repeated NO_2 measurements were carried out in 15 homes of employees of the University. In all homes gas was used for cooking; 3 homes were equipped with an unvented geiser in the kitchen. Another 3 homes had a vented geiser. The repeated RSP and VOC measurements were carried out over a period of half a year in a subsample of 4 of the 15 homes; 2 of these homes had smoking occupants. Since the main objective of the repeated measurement program was to assess the variability of the indoor pollutant levels, a detailed description of the homes and occupants is not considered useful. The homes could be best described as (upper-)middle class homes.

2.4. Summary and discussion

Real-time monitoring program

In the 12 homes of the real-time monitoring program the use of the cooking range of 2 to 6 times per day was typical. The geiser was used 10 to 35 times a day. In 1 home the oven was used for additional space heating. Judged on the data plots, the pattern of use of the gas appliances in each home was more or less stable from day to day. This is in agreement with the results reported by Dijkhof and Ogink (1978). In their study in 38 homes, little differences were observed in frequency and duration of use of gas appliances on different days of the week.
Dijkhof and Ogink reported an average frequency of use of the geiser of 31 times per day, which is higher than observed in the homes of the real-time monitoring program. The presentation of the results for the cooking range by Dijkhof and Ogink do not allow a comparison with the results of the real-time monitoring program.

In the tracer gas experiments in the kitchens, large short-term fluctuations in the tracer gas concentrations were observed, indicating the dynamic and complicated nature of air mixing and -transport in occupied homes. Large differences in 4-hour average ACR and TI were found within homes and between different homes. The overall mean ACR over the entire measurement period in each home ranged from 0.5 to 9 air changes per hour. Although these values are not unrealistic and in good agreement with results of initial tests with the tracer gas experiments (de Haas 1982), the ACR may be seriously affected by deviations of the effective ventilation volume from the physical volume of the kitchen.

Week-long measurement program

The positive response in the week-long measurement program was satisfactory, considering that occupants had no direct gain from participating in the study. Many occupants thought that air pollution was an outdoor phenomenon, but volunteered nonetheless. More than 50 % of the occupants which could be contacted agreed to cooperate. The contact rate in Rotterdam, however, was low, among others because of city renewal and renovation activities in the study area. Households with all members working full-time were probably underrepresented, because they had less chance to be contacted, then households with members who were at home during office hours.
Large differences between the 3 age-groups of homes were found with respect to the distribution of general characteristics of the homes.

As far as possible the distribution in the 3 age-groups of homes of characteristics like home ownership, building type, cooking fuel, type of water- and space heater, was compared with data of the municipality of Ede and various other sources for similar groups of homes. These comparisons revealed no serious deviations.

In the use of ventilation provisions, a more or less bimodel distribution was observed, with one group of occupants who practiced airing and used their ventilation provisions during relatively short periods of time, while others had the ventilation provisions open almost continuously. In general, the ventilation provisions were used over shorter time periods in Rotterdam than in Ede.
During the heating season the ventilation provisions in the living room were never used in about 15 % to 20 % of the homes, according to the questionnaire; for kitchens this was the case in 5 % of the homes. Ventilation provisions in bedrooms were never used in 3 % of the homes in Ede and in 15 % of the homes in Rotterdam. Van Beek et al. (1981) reported that in 8 % of the 1500 homes in their study ventilation provisions in the living room were not used. It is not clear, however, to which season this figure applies. Questionnaire information about ventilation habits during the summer, in the homes of the week-long measurement program, indicated that, as could be expected, occupants tend to increase the frequency and duration of use of the ventilation provisions in the summer, as compared to the winter.
Recently, van Dongen (1984) has published the results of a study in 36 newly built terraced homes, in which the ventilation habits were investigated using diaries. He observed that on average the ventilation provisions in the bedrooms were used during about 25 % of the day. In Ede and Rotterdam the median of the use of ventilation provisions in bedrooms was 42 % respectively 7 % of the time. The differences between the average use of ventilation provisions in Ede, Rotterdam and in the homes studied by van Dongen, in addition to the wide range in the use of ventilation provisions observed in Ede and Rotterdam, suggest that generalizations about typical ventilation conditions have little meaning as long as the motives for ventilation behaviour are not understood.

The gas cooker was used on average during about 1 hour per day in both towns according to the diaries. Dijkhof and Ogink (1978) reported that the most frequently used burners of the cooking range were operated about 40 to 50 minutes per day. This is in good agreement with the results of the week-long measurement program, assuming that several burners are used simultaneously for the preparation of hot meals.
For geisers Dijkhof and Ogink reported an average use of 28 minutes per

day. Since only the prolonged use of the geisers was recorded in the diaries, a comparison of the diary results with the results of Dijkhof and Ogink is not well possible.
Just as for ventilation provisions, a wide range was observed in the use of gas appliances in Ede and Rotterdam. Predictions of indoor pollutant levels based on "typical" gas appliances use, like was done by Welch (1982) and Palsma (1982), have therefore little value for the prediction of the distribution of exposures to gas-combustion related indoor pollutants.

The average tobacco consumption in the homes in Ede and Rotterdam was about 5 cigarettes per day; the data of the questionnaire and diaries were in good agreement with each other. The number of smokers in the home explained about 55 % of the variance in the (logarithm) of the tobacco consumption.
Most studies on tobacco consumption adress the consumption of individual smokers, which does not necessarily takes place inside the homes. A comparison of the results with other studies is therefore not possible.

The use of 8 different groups of possible indoor sources of VOC could be identified from the questionnaire information. The most widely used products were aerosol sprays, which were used in about 50 % of the homes during the measurement period. The classification of product use was crude however, because a quantification of the amount of products which was used was not possible. Reference material to evaluate the results of the use of VOC sources was not available.

The results of the tracer gas experiments revealed a large range in ACR and TI in kitchens and living rooms in both towns. It is obvious both from the real-time monitoring program and from the week-long measurement program that the effective ventilation volume of a room may seriously deviate from the physical volume. This affects the value of the ACR in an unknown manner.
The TI is a parameter, the value of which does not involve the room volume. It directly describes the efficiency in protecting the sampling point from exposure to the tracer gas, no matter how complex the ventilation system is. The usefulness of the TI to evaluate ventilation, however, depends heavily on the location of the tracer gas emission and sampling points. Careful selection of these points, made possible by flexible emission and sampling tubing, is therefore required.
Duplicate SF_6 samples with a coefficient of variation of 15 % taken in 5 kitchens, 10 living rooms and 14 open kitchens/living rooms in the week-long measurement program, showed that differences in the location of the sampling points had only a minor effect on the outcome of the TI.
There is little reference material available for a comparison of the ob-

served long-term average ACR and TI in occupied homes. Turner et al. (1984) recently published the results of long-term tracer gas experiments in occupied homes in the U.S. However, the ACR was expressed for the whole house, assuming one single compartment. The differences in the TI in kitchens and living rooms signify that in the Netherlands, most homes cannot be considered as one single compartment. Neither can homes be considered as a fixed conglomerate of several compartments, as can be judged from the data on the use of interior doors in the week-long measurement program.

CHAPTER 3. RESULTS; CARBON MONOXIDE

In this chapter the results of the real-time monitoring program and week-long measurement program will be presented for CO. CO measurements were not included in the repeated measurement program. In addition, results will be discussed of an investigation of the CO emission of geisers and of a study on CO in exhaled breath as an indicator for indoor CO exposure.

3.1. Real-time monitoring program

For a qualitative examination of the data the CO concentrations in each location in each home were plotted versus time of day. In most homes elevated CO levels coincided with the use of the gas cooker. Elevated CO levels were also observed during the use of unvented geisers.
In home 1, 2, 3, 8 and 10, the contribution of the unvented geiser to the CO level in the kitchen was less than that of the gas cooker. The geisers in these homes were equipped with secundary aerated burners. The effect of unvented geisers on the kitchen CO levels seemed more pronounced in home 6, 11 and 12. These geisers had primary aerated burners. Peak concentrations of CO were about the same during the use of gas cooker and during the use of the unvented geiser in these homes.
These observations however, are only tentative and based on qualitative examination of data plots. In most cases during periods of use of the gas cooker, also the geiser was frequently operated. This, in addititon to the limited number of homes and the many differences between homes, precluded a clear distinction between the impact of the cooker and that of the unvented geiser on kitchen CO levels.

Figure 7 shows the plots of the CO level versus time of day in 3 locations in home 12. The figure shows sharp peaks in the CO level in the kitchen due to the use of gas appliances. These peaks were reflected in the pattern of the CO level in living room and bedroom, but in a smoother form. CO levels in these locations remained lower than in the kitchen. Similar concentration patterns were observed on other days and in other homes as well. In some homes differences were observed in the air communication between the locations on different days. In these homes the CO concentration patterns in the living room and bedroom corresponded well with that in the kitchen, while on other days the CO concentrations in these rooms were hardly affected by high CO levels in the kitchen.
In the living room of homes of smokers slightly elevated CO levels of a few

mg/m^3 were observed during periods when gas appliances were not used. These patterns suggest a moderate impact of tobacco consumption on indoor CO levels. An example of such a pattern is shown in figure 7 (evening hours).

For the 3 indoor locations in each home 1-minute and 1-hour average CO concentrations were calculated and the overall mean CO concentration over the entire measurement period was determined. The frequency distribution of the 1-hour average CO levels in the indoor locations in each home is given in figure 8. This figure also contains the maximum 1-minute average and 1-hour average CO concentration and the overall mean CO concentration in each home. The range in the maxima of short-term average CO concentrations and in the overall mean concentration are summarized in table 19.
The overall mean CO level in the indoor locations was 0 to 3 mg/m^3 higher than the outdoor level. Differences between indoor locations and between different homes were small (less than 3 mg/m^3).
When the 1-minute average and 1-hour average CO levels of different homes were compared, there appeared to be more differences than simularities. In some homes the differences between the maximum 1-minute average and 1-hour average CO concentrations were large (in kitchens sometimes tens of mg/m^3) while in other homes the differences were only a few mg/m^3. Differences in CO levels between locations were also small in some homes but large in others.
Kitchen concentrations were generally higher than the concentrations in other locations. In home 5 the highest CO levels were observed in the bedroom. Examination of the data plots learned that elevated CO levels in the bedroom were not typical in home 5. This single high value of 48 mg/m^3 (1-minute average CO level) coincided neither with elevated levels in the other locations nor with the use of gas appliances.
This high CO level might been caused by a puff of tobacco smoke near the sampling point in the bedroom.
Examination of the plots indicated that the relatively frequent occurrence of 1-hour average CO concentrations in the range of 2 to 6 mg/m^3 in the living rooms of home 1, 4, 5, 6, 11 and 12, were probably the result of tobacco smoking. In home 6 and 11, however, an effect of (vented) gas-fired space heaters on the CO levels in the living room cannot be excluded.

According to the proposed air quality standard for ambient CO levels, a 1-hour average value of 40 mg/m^3 should not be exceeded more than once a week and an 8-hour average value of 10 mg/m^3 should not be exceeded more than once a month (Dutch Health Council 1979). The 1-hour average value of 40 mg/m^3 was exceeded once in the kitchens of home 6 and 11; in home 11 the 8-hour average value of 10 mg/m^3 was also exceeded once.

3.2. Week-long measurement program

Acceptable CO measurements were obtained in 69 kitchens and 68 living rooms in Ede and in 45 respectively 51 kitchens and living rooms in Rotterdam. The frequency distribution of the weekly average CO concentrations are given in figure 9.
Typical outdoor CO levels, obtained from the nearest station of the National Air Pollution Monitoring Network and in addition in Ede from occasional outdoor measurements with a CO Ecolyser, were 1 mg/m^3 or less during both winters.
The weekly average CO concentrations in the 2 towns were comparable to the overall mean CO levels that were found in the 12 homes of the real-time monitoring program.
In Rotterdam weekly average CO levels of 1 mg/m^3 or less were observed more frequently in the kitchens than in Ede. The maximum CO concentration in kitchen and living room of 9 mg/m^3 were found in the same home in Rotterdam. This home had an open connection between kitchen and living room.

The possible associations between indoor CO levels and characteristics of the homes and its occupants were tested by multiple regression analysis (cf. part 1, section 3.6).
The logarithm of the CO levels in the kitchen were regressed on the independent variables in table 20. These independent variables were selected for the regression analysis, because a potential effect on indoor CO levels was reported in the literature or because an effect had been found in related studies. The independent variables were devided in 2 sets: one which represented the presence of indoor CO sources and ventilation appliances and another, more detailed set, which also incorporated the use of CO sources and ventilation provisions and the tracer gas transfer in the kitchens.
The variables 2 to 4 which were related to the presence of a geiser were entered in the regression analysis as dummy variables.
The variables in set 2 obtained from the diaries were set to a value of 0 when an appliance was not present in the home. Note that the number of cases, with a complete data set for all variables in the equation, is small, especially in set 2.
Because of the small number of cases, the independent variables were selected in the regression equation at a significance level of < 0.10, instead of 0.05. The regression equations derived from the analysis with the variables of set 1 is given in table 21. In equation 1 in table 21 the burner type of the geiser was represented by 2 dummy variables, in equation 2 only by 1 dummy variable for the secundary aerated burners.
Only 15 % of the variance in the logarithm of the kitchen CO level could be

explained by the selected independent variables.
The variables 'unvented geiser' and 'secundary aerated burner' had no significant effect on the dependent variable after the variable 'primary aerated burner' was selected in the regression equation 1. In equation 2, the effect of the variable 'unvented geiser' was almost completely undone by the variable 'secondary aerated burner' as can be judged from the regression coefficients of these variables. These results indicate that the effect of unvented geisers on the kitchen CO levels is mainly caused by appliances equipped with primary aerated burners.
Both equations showed that the presence of smokers in the home was positively associated with the CO level in the kitchen.

The results of the regression analysis with the independent variables of set 2 are presented in table 22.
The variables 'city', 'use of geiser pilot light' and 'use of gas cooker' were selected in the regression equation and explained 18 % of the variance in the logarithm of the CO levels in the kitchen. To test whether the variable 'use of geiser pilot light' was only a proxy for the presence of unvented geisers, the regression equation was recalculated after adding the variable 'presence of unvented geiser' and a dummy variable for 'secundary aerated burner' to the independent variables of set 2.
Indeed the effect of the variable 'use of geiser pilot light' on the kitchen CO levels disappeared after these variables were added (table 23).
In the equation in table 23 also a positive association between tobacco consumption and the CO level was observed. The variable 'tobacco consumption' only reached borderline significance ($p = 0.11$) in the equation in table 22.

To explain the observed differences in the weekly average CO levels in the living room between homes, the logarithm of the living room CO level was regressed on 'the logarithm of the CO level in the kitchen', 'the number of doors between kitchen and living room', 'the presence of smokers' and 'city' as independent variables. The equation which resulted from this regression analysis is given in table 24.
No statistically significant association was found between the variable 'city' and the CO level in the living room. The remaining 3 independent variables explained 34 % of the variance in the logarithm of the CO level in the living room.
Additional regression analysis, incorporating also the use of interior doors and of ventilation provisions, the tobacco consumption and the index of tracer gas transfer from the kitchen to the living room, did not contribute to a better explanation of the CO levels in the living room. In the additional analysis it was observed that the variable 'tracer gas transfer

index in the living room' had about the same effect on the CO levels in the living room as the variable 'number of doors between kitchen and living room'.

3.3. Related studies on indoor CO pollution

Potential CO emission of geisers

To assess the potential CO emission of geisers, a survey type of study was carried out in November, December 1980 in the towns of Arnhem and Enschede (Brunekreef et al. 1982). Enschede was selected because a special maintenance system for geisers was practiced in this town.
Arnhem was considered to be more typical for the situation in the Netherlands with regard to geiser maintenance.
The investigation covered 254 homes that came from a random sample of 815 (537 in Arnhem, 278 in Enschede), out of all homes that 'most probably' were equipped with a geiser.
In each home the air pollution potential of the geiser was assessed by operating the appliance continuously for 15 min. The CO concentration in flue gases was measured after 1 and 15 min. of operation, the CO concentration at breathing height (1.5 m) was measured before and after the 15 min. period. All measurements were taken with windows and doors closed. Permanent ventilation ducts, however, were left open. The installation code requires that gas appliances function properly under these circumstances.
The CO measurements were performed with Ecolyzer 2000 monitors.
In addition to the measurements, a number of items were recorded by the investigators including: age and burner type of the geiser, presence of a flue and geiser maintenance.

The results of the CO measurements were expressed in ppm (1 ppm CO = 1.15 mg/m^3).
In 27 % of the investigated homes, CO concentrations in flue gases exceeded requirements of the local gas companies (300 ppm). The main parameter determining CO concentrations in flue gases was the type of burner. Of all the primary aerated burners, 32 % reached CO concentrations higher than 300 ppm. In case of secondary aerated burners this was only 3 %. For the group of geisers with a primary aerated burner, the maintenance system appeared to be an important factor to prevent high CO concentrations in flue gases.
In 17 % of cases, CO concentrations at breathing height exceeded a value of 50 ppm, the installation code standard. In some cases a level of more than 600 ppm was observed. Presence of a vent and, again, type of burner both had an effect on CO levels at breathing height. In 3 % of the cases, vented

geisers produced CO concentrations higher than 25 ppm, while in 50 % of the homes in which unvented geisers with a primary aerated burner were located, CO levels exceeded 25 ppm. In 12 % of the homes equipped with an unvented geiser with a secundary aerated burner, CO levels exceeded 25 ppm. In addition to the burner type and presence of a vent, the maintenance system proved to be important in reducing CO levels at breathing height.

CO in exhaled breath

To establish the health consequences of geisers with a high CO production under normal living conditions, CO levels in exhaled breath were determined from 29 occupants (age 12-72 years) of 15 flats (Verhoeff et al. 1983). The CO level in exhaled breath is directly related to the carboxyhemoglobin (COHb) level in the blood and can be used as a measure for the exposure to CO. COHb levels below 2.5 to 3 % are considered safe. Above this level, health effects have been observed in sensitive individuals (WHO 1979b).

All flats were identical and equipped with an unvented geiser with a primary aerated burner, which had a CO concentration in flue gases of over 250 ppm. Duplicate samples of exhaled breath were taken on 2 days, before and after the cooking and dish washing period, during which the frequent use of the geiser is likely to occur. After holding their breath for approximately 20 seconds, participants exhaled through a glass tube of 225 ml. After exhalation the tubes were closed and samples were analysed on CO content gaschromatographically at the laboratory. The overall coefficient of variation of series of duplicate samples was about 7 %. COHb levels were calculated from the CO concentrations in the samples using the equations of Ringold et al. (1962) and Peterson (1970).

Small, but in some cases statistically significant, increases in CO levels in exhaled breath of about 1 ppm were found in both smokers and non-smokers after the cooking/dish washing period. For the smokers this could also be due to increased tobacco consumption during dinner time. An association between the CO concentration in flue gases and the increase of CO-breath levels was not found.
In non-smokers COHb levels remained below 2.5 % before as well as after the cooking/dish washing period. The majority of the smokers had COHb levels of over 2.5 % both before and after cooking/dish washing. The rank-order correlation between average daily tobacco consumption and COHb levels before cooking/dish washing in 15 smokers was 0.78 ($p < 0.01$).

The results of the study indicated that apart from the CO production of the

geiser, other factors like kitchen ventilation, presence or absence of occupants in the kitchen during appliance use, etc. play a crucial role. The frequency of occurrence of biologically significant increases in COHb levels due to geisers with a high CO production is thus presumably much lower than the frequency of occurrence of geisers with a high CO production itself. Identification of a specific population at risk, by means of a general survey, is therefore probably unfeasible.

3.4. Summary and discussion

In the homes of the real-time monitoring program, 1-hour average peak concentrations of CO of a few mg/m^3 and in some homes of a few tens of mg/m^3 were observed in the kitchen and in the living room. With the exception of 1 home, in which 1-hour average CO concentrations of over 25 mg/m^3 were observed in the bedroom, bedroom CO levels were only a few mg/m^3 higher than outdoor levels.
In 1 home a 1-minute average peak concentration of over 100 mg/m^3 CO was measured in the kitchen. Examination of the data plots learned that CO peak concentrations in the kitchen coincided with the use of unvented gas appliances. The data were not considered conclusive, due to the limited number of homes and the many differences between homes, to discriminate between the impact of different types of unvented gas appliances on the CO levels in the kitchen.
From the data plots and from the relatively frequent occurrence of 1-hour average CO levels in the range of 2-6 mg/m^3 in the living rooms of homes of smokers it was inferred that tobacco consumption had a moderate effect of a few mg/m^3 on the CO levels in the living room.

Similar CO levels as were found in the real-time monitoring program, have been reported in other countries for normal occupied homes equipped with unvented gas appliances (Elkins et al. 1974, Wade et al. 1975, Moschandreas et al. 1978, Sterling and Sterling 1979, Hawthorn et al. 1983, Viala et al. 1983).
The small contribution of tobacco smoke to the CO levels in the living room is in good agreement with results published by Harke et al. (1974), Sebben (1977), Fisher et al. (1978) and Grandjean et al. (1979).

The value of 40 mg/m^3 CO (1-hour average) of the proposed standard for ambient CO was exceeded in the kitchen of 2 homes; the 8-hour average value of 10 mg/m^3 was exceeded in the kitchen of 1 home.The proposed standard was not exceeded in living rooms and bedrooms.
In 1980, the US Environmental Protection Agency (EPA) proposed to lower the

1-hour average standard for CO to 25 ppm (29 mg/m^3). This value was almost reached in the living room and bedroom of 1 home, where the highest 1-hour average CO concentrations were 26 mg/m^3. According to the installation code for gas appliances (NEN 1078, 1978) the CO level should not exceed a value of 50 ppm (58 mg/m^3). This code, however, is of little value since it does not specify the time period to which the value of 50 ppm applies.
The Working Group on Indoor Climate of the Dutch Health Council has recently expressed the view that the values of the proposed air quality standard for CO should preferably be lower for indoor situations than for outdoor air(Dutch Health Council 1984).
In a strict sense, the duration of the measurement period in each home was too short and the number of homes was too small to conclude whether the proposed standard for ambient CO was exceeded or not. The data do warrant the conclusion that, if any, the margin of safety between the proposed standard and the measured CO levels in some homes will be very small.

In the homes of the week-long measurement program, the weekly average CO levels were similar to the overall mean CO levels found in the 12 homes of the real-time monitoring program.
On average, the weekly average CO level was 2 mg/m^3 in the kitchen and 1 mg/m^3 in the living room in both towns.
The multiple regression analysis with the logarithm of the indoor CO levels showed that the presence of unvented geisers equipped with primary aerated burners, smoking of tobacco and the duration of use of the gas cooker were positively associated with the CO levels in the kitchen. Contrary to the expectations, the CO levels in kitchens in Rotterdam were lower than those in Ede, even after standardisation for the above mentioned independent variables. The reasons for these differences between the 2 towns are not clear, but may be caused by differences in kitchen geometry, appliance use and appliance maintenance system.
The weekly average CO levels in the living room were positively associated with the CO level in the kitchen and the presence of smokers. The negative association of the number of doors between kitchen and living room with the CO level in the living room indicated that these doors are effective in reducing the transfer of CO from the kitchen to the living room.

The effect of unvented geisers and burner type of the geiser on CO levels in the kitchen, was also previously found as important determinant of kitchen CO levels during the use of the geiser under standerdized conditions. After 15 minutes of operation of the geisers, CO levels of several hundreds of mg/^{3} were observed in some homes. The results of the study on CO emissions of geisers indicated that, although presence of a vent and a secundary aerated burner were clearly associated with lower CO levels,

these factors are no guarantee that kitchen CO levels will not exceed 25 ppm. In addition to the presence of a vent and the burner type, the maintenance system proved to be an important factor for CO levels in the kitchens under standardized conditions.

Under normal living conditions only slight, but in some cases statistical significant, increases in COHb levels were found after the cooking and dish washing period in occupants of homes equipped with unvented geisers with a high CO production. The results of the study on COHb levels indicated that, under normal living conditions, the frequency of occurrence of biological significant increases in COHb levels due to the use of geisers is less than the frequency in which geisers with a high CO production itself are observed in the general housing stock.

CHAPTER 4. RESULTS; NITROGEN DIOXIDE

In this chapter the results of respectively the real-time monitoring program, the week-long measurement and the repeated measurement program will be presented. Furthermore, the results of week-long NO_2 measurements from 3 related studies will be reported.

4.1. Real-time monitoring program

Just as for CO, the results of the NO_2 measurements were first plotted for a visual inspection of the data.
Clearly elevated NO_2 levels were observed in the kitchen during the use of the gas cooker and even more during the use of the unvented geiser. An example of the plots of the use of gas appliances and the simultaneous NO_x concentration in the kitchen of home 2 is given in figure 10.
During the use of the geiser the ratio of NO to NO_2 was about 2. The ratio of 2 was typical for other homes with unvented geisers as well. The ratio of NO to NO_2 during the use of the gas cooker was less constant than during the use of the geiser. Figure 11 shows the NO_x concentration versus time of day in the 3 indoor locations of home 9, where the gas cooker was the only gas appliance. In the kitchen the ratio of NO to NO_2 during the use of the cooker varied between about 1 and 2. This was probably the result of differences in fuel input and cooking load.
The NO_x concentration patterns in the living room and bedroom were generally smoother and peak concentrations remained lower than in the kitchen. In the example given in figure 11, the NO_x level in the bedroom was hardly affected by the NO_x levels in the kitchen.
There were no indications of tobacco smoke having an effect on the NO_2 levels. On several occasions it was observed that in the homes of smokers, the NO level in the living room exhibited the same pattern as the CO concentration during periods when gas appliances were not used. This suggests that tobacco smoke might elevate NO levels.
Following the same reasoning as for CO, it should be noted that the observations from the qualitative examination of the data plots on the ratio of NO to NO_2 during the use of gas appliances and on the impact of tobacco smoke on the NO_x levels have a tentative character.

For the NO_2 concentrations in the 3 indoor locations, 1-minute average, 1-hour average and 24-hour average values were calculated and the overall mean concentration over the entire measurement period in each home was

determined. The frequency distribution of hourly average NO_2 levels in the 12 homes is given in figure 12. In addition, figure 12 contains the maximum 1-minute, 1-hour and 24-hour average concentration observed in each home. The range of these values over the 12 homes is summarized in table 25.
Especially in the kitchen extremely high 1-minute average NO_2 concentations of several hundreds up to over 3500 $\mu g/m^3$ were observed. The highest 1-hour average NO_2 level was about 2000 $\mu g/m^3$ (home 11). In the other homes, the maximum 1-hour average NO_2 levels were less than 1000 $\mu g/m^3$. Living room and bedroom NO_2 levels were generally lower than the levels in the kitchen. 1-Hour average NO_2 levels of several hundreds of $\mu g/m^3$ were observed in living rooms and bedrooms of some of the homes.
In 4 homes (3, 4, 5 and 7) the maximum 1-hour average NO_2 level in the living room was about 15 $\mu g/m^3$ higher than that in the kitchen.
In home 3 the maximum 1-hour average NO_2 level in the living room exceeded the level in the kitchen on a day when an almost uniform distribution of CO, NO and NO_2 levels was observed in the kitchen and in the adjacent living room and bedroom. Figure 13 shows the plots of the NO_x level versus time of day in the 3 indoor locations. During a period from about 9.30 h. till 13.00 h. the gas cooker was used continuously and the unvented geiser occasionally. On most other days the concentration patterns in this home were smoother and peak concentrations were lower in the living room and bedroom than in the kitchen.
Home 5 and 7 had an open kitchen/living room.
The higher maximum 1-hour average NO_2 level in the living room than in the kitchen observed in home 3, 5 and 7, were probably the result of the sampling sequence in the real-time monitoring program. The time lag of about 2 minutes between the samples in the indoor locations introduced small differences in the time periods over which the 1-hour average NO_2 concentrations were calculated. Given the observed concentration patterns, these time lags may account for the small differences in the 1-hour average NO_2 levels in different locations.

No satisfying explanation was found for the high 1-hour average NO_2 concentration of 334 $\mu g/m^3$ in the living room of home 4. This value was measured at a time when the cooker and vented geiser were not used. Figure 14 shows a plot of the NO_x concentrations in the living room on that day. After about 14.00 h., the NO_2 concentration in the living room started to rise gradually, while NO and CO levels remained the same. NO_2 levels in the kitchen and outdoors were not elevated, while the bedroom NO_2 level reached a level of 150 $\mu g/m^3$. At 16.35 h., the NO_2 level in the living room sharply declined. This decline might have been caused by occupants activities, because at the same time the gas cooker and geiser were used, which elevated NO and CO levels in the kitchen and the living room. A similar, but

less pronounced built-up of NO_2 was observed on 2 other occasions in home 4: once in the living room between 14.00 h and 17.00 h. and once in the kitchen between 10.00 h. and 12.00 h.. On the latter occasion, the NO_2 concentration pattern in the kitchen was reflected to some extent in the living room. Backflow of chimney gases from the space heater and from the geiser might be an explanation for this phenomenon, although in that case the NO level would probably be elevated as well, unless NO would be completely transformed in NO_2 during the contact with chimney surface. Interferences from another unrecognized pollutant generated indoors might also be an explanation for the observed concentration patterns. The anomalous NO_2 concentrations were excluded from further data analysis.

Comparison of indoor NO_2 levels with the air quality standard for outdoor NO_2.

As mentioned in part 1,section 1.4, the Dutch Health Council has proposed air quality standard for outdoor NO_2 levels. The proposed standard reads as follows: an 1-hour average NO_2 concentration of 135 $\mu g/m^3$ is not to be exceeded during 98 % of the year, while a value of 300 $\mu g/m^3$ is not to be exceeded more than once a year; a 24-hour average NO_2 concentration of 120 $\mu g/m^3$ is not to be exceeded during 98 % of the days, while a value of 150 $\mu g/m^3$ is not to be exceeded more than once a year. The standard is aimed at the prevention of short-term peak concentrations. The 1-hour average value of 300 $\mu g/m^3$ is considered as a maximum; the other values were derived from this maximum, based on the frequency distribution of the hourly and daily NO_2 concentrations under the prevailing meteorological conditions of the outdoor atmosphere.
Since the frequency distribution of NO_2 concentrations in homes is quite different from that in outdoor air, only the 1-hour average value of 300 $\mu g/m^3$ is suitable for evaluation of indoor levels. Table 26 presents the results of the comparison between 1-hour average NO_2 levels in the 12 homes and the value of 300 $\mu g/m^3$. In only 3 of the 12 homes the value of 300 $\mu g/m^3$ was not exceeded in either of the indoor locations. In the kitchen of the other 9 homes this value was exceeded during 2 to 22 % of the measurement period. The level of 300 $\mu g/m^3$ was exceeded in the living room and bedroom in 6 respectively 3 homes.

Day to day variation of indoor NO_2 levels.

To assess the daily variability of indoor NO_2 levels, data were used from 7 homes in which succesful NO_2 were obtained in all 3 indoor locations over 7 consecutive days. The variability of the indoor NO_2 levels was expressed as

the reliability coefficient (RC, cf. part 1, section 3.6). The RC was calculated for the daily maximum 1-hour average concentration and for the 24-hour average concentration, using the 'Reliability' routine of SPSS. RC were calculated as an index for the reliability of measurements of one single day, and also as an index for the reliability of the average values over 7 days.
The calculated RC are presented in table 27. The RC for a single day showed that about 50 % (in the living room more than 60 %) of the variance in the daily maxima of 1-hour average concentrations can be considered as random or error variance. This indicates a considerable day to day variation in the peak-concentrations within homes.
The day to day variation of 24-hour average concentrations within homes was smaller; about 30 % of the variance could be considered as error variance. The RC calculated for the average values over 7 days were high. About 90 % of the variance in the average values could be considered as 'true' variance.

The relationship between short-term and long-term NO_2 concentrations.

The relation between short-term and long-term average NO2 concentrations in the 3 locations in the homes of the real-time monitoring program was established in a regression analysis. This was done by calculating the squared multiple correlation coefficient (R^2) and the coefficient of alienation (CA) of the overall mean NO_2 concentration with the maximum 1-minute and 1-hour average concentration in each home. In this analysis, the overall mean concentration is regarded as a predictor for the maximum 1-minute and 1-hour average NO_2 concentration in the homes. Thus, the R^2 expresses the percentage of variance in the short-term concentration peaks which is explained by the overall mean concentration. The CA expresses the gain in the accuracy in predicting the short-term concentration peaks which arises from using the knowledge of the predictor variable, i.e. the overall mean concentration (cf. part 1, section 3.6).
The results of the analysis are presented in table 28. In all cases, more than 50 % of the variance in the short-term concentrations could be explained by the overall mean NO_2 concentration. Yet the gain in accuracy by using the overall mean concentration in the kitchen to predict short-term concentration peaks, is only 30 %. The accuracy of the prediction of the 1-hour average peak-levels in the living room, using knowledge of the overall mean concentration, was 50 % better than it would be without this knowledge. In the bedroom the gain in accuracy was even better.
The accuracy of the prediction of the maximum 1-hour average concentration, expressed in concentration units (the standard error of estimate,cf. part 1

section 3.6), was 346 $\mu g/m^3$ in the kitchen, 129 $\mu g/m^3$ in the living room, and 74 $\mu g/m^3$ in the bedroom.
The ratio of the maximum 1-hour peak concentration to the overall mean NO_2 concentration, the peak-to-mean ratio, estimated by the slope of the regression line, was on average 6 in the kitchen (range 2.7-19.8) and living room(range 2.5-7.9), and 4 in the bedroom(range 2.0-6.9).

4.2. Week-long measurement program.

The results of the week-long measurements in Ede and Rotterdam are presented in figure 15. This figure shows the frequency distributiom of the weekly average NO_2 concentrations in the 3 indoor locations, and in addition, the geometric mean and maximum NO_2 concentration in the 2 towns. The figure also shows the mean outdoor NO_2 level during the measurement period in Ede and Rotterdam.
The weekly average NO_2 levels in the living rooms and bedrooms in the prewar homes in Rotterdam were on average about 10 $\mu g/m^3$ higher than those in the post-war homes of Ede. In the kitchens,NO_2 levels were on average 20 $\mu g/m^3$ higher in Rotterdam than in Ede. In a majority of the homes, kitchen NO_2 levels exceeded outdoor levels. Outdoor NO_2 levels were frequently exceeded in the living room and,to a lesser extent, in the bedrooms. In the week-long measurement program, the maximum weekly-average NO_2 level in the kitchen was about 300 $\mu g/m^3$ higher than the highest overall mean NO^2 concentration which was observed in the kitchens of homes in the real-time monitoring program.

Comparison of indoor NO_2 levels with the air quality standard for outdoor air.

As mentioned before, only the 1-hour average value (300 $\mu g/m^3$) of the proposed air quality standard for outdoor NO_2 is appropriate to evaluate indoor NO_2 levels. To compare weekly average NO_2 levels with the 1-hour average value of 300 $\mu g/m^3$, assumptions had to be made about the ratio of 1-hour average peak-concentrations to the weekly average NO_2 levels. For this purpose, the peak-to-mean ratios,obtained in the real-time monitoring program were used, as the best available estimates. It should be noted, however, that these ratios were determined in a limited number of homes and substantial deviations are likely to occur in other homes, especially in inner-city homes in Rotterdam, where the geometry of the homes was quite different from that of the homes of the real-time monitoring program.
Table 29 presents the percentage of homes in which the value of 300 $\mu g/m^3$

was exceeded, assuming a peak-to-mean ratio of 6 in the kitchen and living room and a ratio of 4 in the bedroom. As could be expected from the frequency distribution of weekly average NO_2 levels (cf. table 15), the value of 300 $\mu g/m^3$ was exceeded more frequently in all indoor locations in Rotterdam than in Ede.

Associations between indoor NO_2 levels and characteristics of the homes and occupants

Just as for CO, the associations between indoor NO_2 levels and characteristics of the home and its occupants were tested by multiple regression analysis(cf. part1, section 3.6). The analysis was carried out on different sets of independent variables for the data of the 2 towns separately as well as combined, using the stepwise selection procedure of the 'New-Regression' routine of SPSS, with a significance level of 0.05.
The indoor NO_2 levels and several of the independent variables were logarithmically transformed for the regression analysis. For convenience, the phrase 'logarithm of' will be deleted throughout the presentation of the results of the regression analysis.
In general, the regression equations for the homes in Rotterdam contained less independent variables then those for the homes in Ede. This was probably due to the smaller number of homes in Rotterdam than in Ede, and due to differences in contrasts in the independent variables between the 2 towns. For instance, in Rotterdam there were only 2 kitchens without a gas cooker, while in Ede 14 homes had no gas cooker in the kitchen.
Although some independent variables in the regression models for the kitchen and the living room in Rotterdam did not reach the significance level of 0.05, the regression coefficients of these variables often had values comparable to those which were selected in the models for the homes in Ede. When the data of the 2 towns were combined, no statistically significant differences between the 2 towns were observed in the NO_2 levels in kitchens and living rooms, after adjustment for the selected independent varaibles. Therefore, only the results of the analysis on the combined data will be presented; differences between the 2 towns will be discussed qualitatively. The regression models for the bedroom will be presented for the data of the 2 towns separately as well as combined.

Kitchen NO_2 levels.

The NO_2 levels in the kitchen were regressed on 3 sets of independent variables (table 30). Set 1 contains simple home characteristics like

presence of unvented gas appliances, and ventilation appliances, while set 2 also incorporated the use of these appliances. Set 3 is an extension of set 2 with the variable 'tracer gas transfer index in the kitchen'. Note that the more detailed information in set 2 and even more so in set 3 was only available for a limited number of homes.
The regression equation resulting from the regression analysis on the variables of set 1, is presented in table 31. In addition to the squared multiple correlation coefficient (R^2), table 31 also shows the coefficient of alienation (CA), as an index for the gain in accuracy which is obtained by using the knowledge of the selected independent variables to predict the dependent variable.
From set 1, 6 independent variables were selected in the regression equation: 'presence of unvented geiser', 'cooking fuel', 'outdoor NO^2 level', 'kitchen volume', 'type of space heating' and 'shower connected to geiser'. Together these variables explained 55 % of the variance in the kitchen NO_2 levels. The CA was 0.68. For the data of the homes in Rotterdam only 'presence of unvented geiser' and 'kitchen volume' were selected in the regression equation. These variables explained 40 % of the variance in the dependent variable.
No statistically significant association was found between the variables 'presence of range hood', 'presence of kitchen window fan' and the NO_2 level in the kitchen. Table 32 illustrates the differences in kitchen NO_2 levels in homes with and without gas cooker and unvented geiser. The combination of presence of an unvented geiser and absence of a gas cooker, however, is rarely observed in Dutch homes.

The results of the analysis on the independent variables in set 2 are presented in table 33. The 7 selected variables explained 61 % of the variance in kitchen NO_2 levels, which is 6 % more than in the equation for set 1. Remarkable in the equation is the positive sign of the regression coefficient of the variable 'use of ventilation provisions in the kitchen', where a negative sign would be expected.
For the data of Ede, a statistically significant negative association was found between the variable 'use of range hood' and the kitchen NO_2 level, after adjustment for the other independent variables in the equation. For the data of Rotterdam, only the variables 'presence of unvented geiser' and 'use of gas cooker' were selected.

The variable 'tracer gas transfer index in the kitchen' of set 3 was not selected in the regression model; from set 3, the same variables were selected as from set 2.

Living room NO_2 levels.

To explain the differences in the NO_2 levels in the living room between homes, a similar procedure was followed as for the NO_2 levels in kitchens. The independent variables which were used in the regression analysis are presented in table 34.
From set 1, the variables 'kitchen NO_2 level', 'number of doors between kitchen and living room' and 'type of space heating' were selected (table 35). The R^2 of the equation was 0.64, the CA was 0.60.
For the data of Rotterdam, the variable 'living room volume' was selected instead of the variable 'type of space heating'. The 'living room volume' was negatively associated with the NO_2 levels in Rotterdam.

The regression equation resulting from the analysis with the variables of set 2 was almost identical to the one for set 1, with only minor differences in regression coefficients (table 36). However, the R^2 of the equation for set 2 was higher than the one for set 1, because, due to missing values, the regression equations were not calculated for identical groups of homes. The use of ventilation provisions and the use of interior doors was not significantly associated with the living room NO_2 levels.

From set 3, the variable 'tracer gas transfer index in the living room' was selected, in addition to the variables already selected from set 1 (table 37). Together, these variables explained 73 % of the variance in the NO_2 levels in the living room. Again, the variable 'living room volume' was selected in the equation for Rotterdam homes, instead of the variables 'tracer gas transfer index in the living room' and 'type of space heating'. When the variable 'kitchen NO_2 level' was removed from the equation as independent variable and only simple home characteristics were used, only 30 % of the variance in the NO_2 levels in the living room could be explained.

Bedroom NO_2 levels.

The regression analysis with the NO_2 levels in the bedroom as dependent variable was carried out on 2 sets of independent variables (table 38). Tracer gas experiments were not carried out in the bedrooms.
In the analysis on the combined data for the 2 towns, a statistically significant difference in the bedroom NO_2 levels was observed between the 2 towns for both sets of independent variables. Therefore, the equations will be presented for the combined data as well as for the 2 towns separately.
The regression models calculated for the variables in set 1 are presented

in table 39. For Ede, Rotterdam and for the combined data, about 50 % of the variance in the dependent variable could be explained. In Rotterdam this was solely due to the association between living room and bedroom NO_2 levels.
Table 40 shows the regression equations from the analysis on the variables of set 2. For the data of the 2 towns combined, the variables 'number of doors between kitchen and bedroom' and 'use of interior bedroom door' were not selected in the model, but reached borderline significance levels of 0.07, respectively 0.08. No significant association was found between use of ventilation provisions in the bedroom and bedroom NO_2 levels.

When only simple home characteristics were used as independent variables and NO_2 levels in other locations were removed from the equation, only about 15 % of the variance in the bedroom NO_2 levels could be explained.

Related studies on weekly average NO_2 levels in Dutch homes.

NO_2 measurements comparable to those in the week-long measurement program were carried out in several related studies (Lebret et al. 1981, Hoek et al. 1984, Noy et al. 1984, Remijn et al. 1984). The results of the NO_2 measurements in these studies are summarized in table 41, in combination with the results of the week-long measurement program. In the above mentioned references, the NO_2 levels were calculated with a different diffusion coefficient for NO_2 than was used in this study (cf. appendix B). For comparison, all the concentration values in table 41 were adjusted to a diffusion coefficient of 0.154 cm^2/m^3. The table shows little differences in the geometric mean NO_2 levels in kitchens and living rooms between rural and suburban areas. In Vlagtwedde, a rural area, bedroom NO_2 levels were on average 10 to 25 $\mu g/m^3$ lower than in the homes located in suburban areas. The geometric mean NO_2 levels in the inner-city homes of Rotterdam were higher in all 3 locations, than in the homes in other areas.

Regression analysis with simple home characteristics as independent variables and NO_2 levels in the kitchen as dependent variable was carried out in several of the related studies. The results were similar to the regression equations which were determined in this study when the variables from set 1 were used. In the related studies, about 40 % to 60 % of the variance in the kitchen NO_2 levels could be explained.

4.3. Repeated measurement program.

The average NO_2 concentrations in the 3 indoor locations of the 15 homes of the repeated measurement program are presented in table 42,broken down by different seasons. Meteorological seasons were defined as follows:winter (1 December - 28 February), spring (1 March - 31 May), summer (1 June - 31 August) and autumn (1 September - 30 November).
In the kitchens, the NO_2 concentrations were highest in the winter and spring , intermediate in autumn and lowest in summer. No clear seasonal patterns were observed in the living room and bedroom. Average NO_2 concentrations in the winter season were about 10 $\mu g/m^3$ less than the average NO_2 concentrations which were measured in Ede in the week-long measurement program.

To assess the variability of indoor NO_2 levels in the 4 seasons, an analysis of variance was performed for each location and each season. Due to holidays, there were a number of missing values in the data, especially in the summer. For the analysis of variance measurement weeks were selected in which there were the least missing values. As a result of this selection, the analysis was performed for 13 homes and 6 weeks in winter, 11 homes and 6 weeks in spring, 11 homes and 3 weeks in summer and for 14 homes and 5 weeks in autumn. The variability of the NO_2 levels was expressed as the reliability coefficient (RC, cf. part 1, section 3.6). Table 43 presents the RC, resulting from the analysis of variance, broken down by location and season. In all 3 locations, the variability of NO_2 within homes was small during winter, spring and autumn, but larger during the summer. Less than 20 % of the observed variance during winter, spring and autumn was error variance according to the RC, indicating that during these seasons indoor NO_2 levels within homes were stable.

4.4. Summary and discussion.

Elevated NO_2 levels were observed in the indoor locations of the homes of the real-time monitoring program during the use of the gas cooker and unvented geiser. NO_2 concentrations were on average higher in kitchens than in living rooms and bedrooms. Concentration patterns were generally smoother in living room and bedroom than in the kitchen. On several occasions it was observed that, just as for CO, NO_2 peak-concentrations in living rooms and bedrooms were virtually the same as the simultaneous kitchen NO_2 levels, while on other days in the same home, levels in living room and bedroom were hardly affected by kitchen NO_2 levels. Due to the pattern of use of the geiser, peak-concentrations were often spread irregularly over

the day.

The ratio of NO to NO_2 in the kitchen was fairly constant during the use of the geiser; during use of the gas cooker, the ratio of NO to NO_2 varied between 1 and 2. These observations were consistent with unreported results of NO_x measurements directly in flue gases of gas cookers and geisers. The observed variation in the ratio of NO to NO_2 during the use of the gas cooker, is in line with results of laboratory studies on pollutant emission rates of gas appliances; the ratio was reported to depend on fuel input, tuning, burning time and cooking load (DeWerth and Himmel 1974, Cote et al. 1974, Billick et al. 1984).
No effect of tobacco smoking on the NO_2 levels could be observed in the homes of smokers.

The 1-hour average peak-concentrations reached levels of several hundreds of $\mu g/m^3$ in all indoor locations in several homes. In 1 home, the highest 1-hour average NO_2 concentration in the kitchen was 2000 $\mu g/m^3$. The observed peak-levels of NO_2 were generally higher than those reported for homes with gas appliances in the USA and the UK (Cote et al. 1974, Moschandreas et al. 1978, Stevenson et al. 1979).
The ratio of the maximum 1-hour average NO_2 concentration to the overall mean concentration, in the homes of the real-time monitoring program, was on average 6 in the kitchen and living room and 4 in the bedroom. With the overall mean NO_2 concentration, more than 50 % of the variance in NO_2 peak-concentrations could be explained. The standard error of estimate, a measure for the accuracy of the prediction of the maximum 1-hour average NO_2 concentration with the overall mean concentration, was over 300 $\mu g/m^3$ in the kitchen, over 100 $\mu g/m^3$ in the living room and about 75 $\mu g/m^3$ in the bedroom. Moschandreas and Zabransky (1982) used similar regression models to predict 1-hour average maximum NO_2 levels from 24-hour average levels measured in 8 kitchens with gas cookers; they reported a peak-to-mean ratio of 2, and a standard error of estimate of 54 $\mu g/m^3$. One of the reasons for the differences between the results of Moschandreas and Zabransky and the results of the real-time monitoring program is probably the effect of unvented geisers on NO_2 peak-concentrations in the Dutch homes. Another reason might be that for the homes of the real-time monitoring program, the peak-to-mean ratio was calculated with the overall mean NO_2 level and not with the 24-hour average NO_2 level. In the homes of the real-time monitoring program, a considerable variation in daily maximum 1-hour average NO_2 concentrations and in 24-hour average NO_2 concentrations was observed within homes. The reliability coefficient of the average value of daily maximum 1-hour average NO_2 concentrations obtained over a period of 7 days, was about 0.90. This indicates that a measurement period of several days is

sufficient to establish differences in indoor 1-hour average NO_2 peak-concentrations between homes.

Also in the homes of the week-long measurement program, indoor NO_2 levels frequently exceeded outdoor NO_2 levels in all 3 locations, but especially in the kitchen. The observed NO_2 levels in Ede and Rotterdam were in good agreement with the results of the related studies on indoor NO_2. In inner-city homes in Rotterdam, the NO_2 levels were generally higher than in the homes in suburban and rural areas.
Comparable or somewhat lower (on average by 10 - 20 $\mu g/m^3$) indoor NO_2 levels than those observed in the week-long measurement program, have been reported for homes with gas appliances in the USA (Cote et al. 1974, Moschandreas et al. 1979, Palmes et al. 1979, Speizer et al. 1979, Spengler et al. 1983). Goldstein et al. (1979) reported mean weekly average NO_2 levels of 210 $\mu g/m^3$ in 428 kitchens equipped with a gas cooker in homes in the UK; this is clearly higher than the geometric mean of the weekly average NO_2 levels which were found in the homes of the week-long measurement program.
In homes equipped with unvented kerosene space heaters in the USA, the UK and Japan, living room NO_2 levels were generally higher than those found in Dutch living rooms. Concentrations reported for the homes with kerosene heaters were comparable to kitchen levels in Dutch homes (Atkins et al. 1979, Nitta and Maeda 1982, Ritchie and Oatman 1983, Leaderer et al. 1984).

Regression models explained about 50 to 70 % of the variance in indoor NO_2 levels between homes, in the week-long measurement program. Goldstein et al. (1979) could account for about 11 % of the variance in kitchen NO_2 levels with similar regression analysis; Spengler et al. (1983) could explain 39 % of the variance in kitchen NO_2 levels and 28 % of the variance in bedroom levels. In homes with and without gas cookers and unvented kerosene space heaters, Leaderer et al. (1984) could account for 60 to 65 % of the variance in indoor NO_2 levels, with regression models incorporating diary results about the use of gas cooker and kerosene heaters.

The regression models for NO_2 levels in the kitchens of Dutch homes showed that the unvented geisers and gas cookers were the dominant NO_2 sources. It is not clear whether the association between type of space heating and NO_2 levels in kitchens and living rooms means that local gas heaters may act as a direct NO_2 source; the variable 'type of space heater' may also be a proxy for other characteristics of the homes or occupants.
No direct association was found between the variable 'use of geiser' and NO_2 levels in the kitchen. However, this variable was positively associated with 'use of gas cooker' and 'use of pilot light of the geiser', which were

both selected in the regression equations and may have incorporated part of the effect of the variable 'use of geiser'. The association of the variable 'shower connection to the geiser' with kitchen NO_2 levels suggests that long use of the geiser is indeed associated with higher kitchen NO_2 levels. This variable is considered to be a proxy for increased use of the geiser; when the geiser supplies hot water to the shower, the geiser is operated over longer periods of time than when it supplies water for other uses (Dijkhof and Ogink 1978). In the homes of the week-long measurement program, the correlation between 'shower connection to the geiser' and 'use of geiser' was 0.75.

The use of kitchen ventilation provisions in the kitchen was found to be positively associated with kitchen NO_2 levels. Apart from chance, this might indicate that higher ventilation enhances the penetration of outdoor NO_2. In that case, however, it would would be more likely to find such an effect of ventilation on the NO_2 levels in the bedroom, since in this location NO_2 levels were generally lower than in the kitchens and in many homes even lower than outdoor levels. Another explanation might be that percieved high pollutant concentrations (for instance water vapor) from unvented gas appliances, stimulate occupants to use the ventilation provisions, but the resulting ventilation increase is not enough to substantially reduce the NO_2 levels. During the field work many respondents mentioned condensation of water vapor in the kitchen as an incentive to use the ventilation provisions.

The only indication for a potential beneficial effect of a range hood on kitchen NO_2 levels, was found in a regression equation for the data of homes in Ede. In 2 of the related studies on indoor NO_2, a similar association was observed between presence/use of range hoods and indoor NO_2 levels.

In the living room, 60 % of the variance in NO_2 levels could be explained by the NO_2 level in the kitchen, the number of doors between kitchen and living room and the type of space heating. The association of the number of doors between kitchen and living room with the living room NO_2 levels, indicates that these doors, or the associated geometry of the home, reduce the transport of NO_2 from the kitchen to the living room.

No significant association was observed between the use of ventilation provisions and interior doors, and the living room NO_2 levels. Neither was there an association between outdoor and living room NO_2 levels. Given the, on average, short periods during which ventilation provisions were used in the living room, it is conceivable that no effect of outdoor NO_2 levels on the indoor levels was observed in the living room, while in kitchens and bedrooms the outdoor level was positively associated with the NO_2 levels in these locations.

The highest proportion of the variance (73 %) in the living room NO_2 levels was explained when the variable 'tracer gas transfer index in the living room' was added to the regression equation. This suggests that after adjustment for the previously mentioned independent variables, differences between homes in the communication of air between kitchen and living room were reflected in the NO_2 levels in the living room.
When kitchen NO_2 levels were not used as explanatory variable, only 30 % of the variance in NO_2 levels could be explained.

The absence of statistically significant differences in kitchen and living room NO_2 levels between Ede and Rotterdam, after adjustment for the independent variables in the regression equations, suggests that the differences in the frequency distribution of NO_2 levels in these locations between the 2 towns were the result of differences in the distribution of indoor NO_2 sources and other factors which influence indoor NO_2 levels. In bedrooms, NO_2 levels in Rotterdam were significantly higher than those in Ede, after adjustment for independent variables. This is probably the result of differences in geometry of the homes between towns. In many homes in Rotterdam, the bedroom and living room were virtually a single compartment,often with the living room area located between kitchen and sleeping area. This geometry was seldom seen in the homes of Ede.

The results of the repeated measurement program showed that in all indoor locations the variability of weekly average NO_2 levels was small during winter, spring and autumn, but larger during summer, when concentrations were lowest. This means that during the greater part of the year, indoor NO_2 levels were consistently different in different homes; with 1 or 2 week-long NO_2 measurements these differences can be detected.
About 15 to 20 % of the variance in indoor NO_2 during winter, spring and autumn could be considered as error variance, due to a.o. measurement errors and differences within homes in occupant activities like use of gas appliances and ventilation provisions. This proportion of the variance can therefore never be explained in regression models with simple home characteristics as independent variables. In the regression equations with detailed diary information, the proportion of unexplained variance was 20 % higher than the error variance of a single week-long NO_2 measurement.

When regression models (developed with the data of the week-long measurement program) are judged on the ability to predict indoor NO_2 (for instance for epidemiological studies), the gain in accuracy of the predictive model is most directly described by the coefficient of alienation, CA. The CA of the regression models showed that the accuracy of the NO_2 levels predicted with regression models was about 30 to 40 % better than a prediction

without such a model. A prediction without a model would mean that the (geometric) mean NO_2 level in Dutch homes would be the best estimate for the NO_2 levels in individual homes, in other words this prediction would be a semi-blind guess.
Since devices for week-long measurements of indoor NO_2 are cheap and easy to operate, direct measurement of indoor NO_2 should be preferred above predictive models in epidemiological studies on the health effects of indoor NO_2. This is not only because obtaining detailed information about characteristics of the home and occupants is extremely laborious, but also because it yields less accurate estimates of indoor NO_2 levels.

Both in the real-time monitoring program and in the week-long measurement program, the 1-hour average maximum value of the proposed air quality standard for outdoor NO_2 was exceeded frequently in the kitchen and regularly in the living room and bedroom. The results of the repeated measurement program suggest that in homes where indoor NO_2 levels exceeded the standard, the exceedance is likely to be recurrent. However, the assumptions about the ratio of peak-to-mean concentrations, needed for the comparison of weekly average NO_2 levels to the 1-hour average value of the air quality standard, were based on measurements in only 12 homes; the error of the prediction of 1-hour average peak-concentrations with weekly average NO_2 concentrations was substantial. This should be kept in mind when interpreting the percentages of homes in the week-long measurement program, in which the proposed air quality standard was exceeded.

At present, it is not clear whether the observed indoor NO_2 levels represent a health risk. The results of studies in this field are inconsistent (Florey et al. 1979, Keller et al. 1979, Melia et al. 1979, Speizer et al. 1980, Comstock et al. 1981, Hasselblad et al.1981, Dodge 1982). In only 1 study, NO_2 was actually measured in the homes of a sufficient number of occupants , to allow a meaningful assessment of the relationship between indoor NO_2 levels and health effect. In the other studies, indoor NO_2 levels were not measured, but predicted from home characteristics. With the present knowledge about the accuracy of such predictions, it is questionnable whether health effects of indoor NO_2 levels would have been detected, if these health effects exist.
Preliminary results of a recent study, in which indoor NO_2 levels were determined in Dutch homes, suggested a negative association between exposure to indoor NO_2 and pulmonary function in non-smoking female occupants, but not in the smoking women in the homes (Fischer et al. 1985). This study was carried out in Vlagtwedde, a rural area in which indoor NO_2 levels were lower than those observed in inner-city homes. The study on pulmonary functions and indoor exposure to NO_2 is now extended to other areas and

population groups; the preliminary results mentioned above need further confirmation from the extended study.

CHAPTER 5. RESULTS; RESPIRABLE SUSPENDED PARTICLES.

In this chapter the results of the RSP measurements in the week-long and repeated measurement program will be presented.RSP was not measured in the real-time monitoring program. Instead, instantaneous RSP measurements were carried out in 3 indoor locations in the homes of the week-long measurement program.

5.1. Week-long measurement program.

Acceptable RSP measurements were obtained in living rooms of 169 homes in Ede and in 91 homes in Rotterdam. The frequency distribution, geometric mean and maximum of the weekly average RSP levels are given in figure 16. Outdoor TSP levels, measured at a central monitoring station of the Central Environmental Protection and Management Agency 'Rijnmond', in Rotterdam were on average 45 $\mu g/m^3$. Background 'Standard Smoke' concentrations determined by the Provincial Department of Environment in Arnhem, the nearest monitoring site to Ede, were on average 25 $\mu g/m^3$. Occasional outdoor RSP measurements with the Piezobalance during the field work for the indoor measurements varied between 15 and 45 $\mu g/m^3$.
In Ede, the geometric mean of the weekly average RSP levels was 5 $\mu g/m^3$ higher than in Rotterdam, the maximum weekly average RSP level in Ede was about 300 $\mu g/m^3$ higher than in Rotterdam, but the differences in RSP levels between the towns were not statistically significant ($p > 0.10$).

As was mentioned in part 1, section 1.4, the Dutch standard for outdoor particulate matter is only applicable for 'Standard Smoke' measurements. Comparison of indoor RSP levels to this standard is therefore not appropriate.
According to the US primary standard for outdoor particulate matter, a 24-hour average TSP concentration of 260 $\mu g/m^3$ should not be exceeded, while the annual geometric mean TSP level should not exceed a value of 75 $\mu g/m^3$. In the proposed revision of the standard, the 24-hour average concentration-limit for particles of less than 10 µm (PM_{10}) ranges between 150 - 250 $\mu g/m^3$, while the annual arithmic mean PM_{10} concentration-limit ranges from 50 - 60 $\mu g/m^3$ (EPA 1984). Table 44 shows the results of a comparison of the weekly average indoor RSP levels to the (proposed) values for 24-hour average outdoor particulate matter.

Associations between indoor RSP levels and characteristics of the homes and occupants.

To explain differences in the observed weekly average RSP levels between homes, multiple regression analysis was carried out in a similar way as for the NO_2 levels. The 3 sets of independent variables which were used in the analysis with the logarithm of the RSP level as dependent variable, are presented in table 45. In the first set, the tobacco consumption indoors is represented by the number of smoking occupants. In the second set, tobacco consumption is represented by questionnaire information about average daily consumption of cigarettes, cigars and pipes in the homes. In the third set, diary information is used as independent variables. The independent variable 'person-hours' in set 3 represents the daily number of people in the home, times the hours these persons spent inside the home during the measurement period. This variable, just like the variable 'family size', is a proxy for indoor activity. As for the regression equations for NO_2 levels in kitchens and living rooms, there were no significant differences in RSP levels between the 2 towns, after adjustment for the independent variables. Therefore only the equations for the combined data of the 2 towns will be presented and differences in the regression equations for the 2 towns separately will be discussed qualitatively.
From set 1, only the variable 'number of smokers' was selected in the regression equation (table 46). This variable explained 40 % of the variance in the RSP levels between homes; the CA was 0.77.
With the questionnaire information from set 2, 50 % of the variance in the dependent variable could be explained by the variables 'cigarette consumption', 'cigar consumption' and 'family size' (table 47).
The regression equation resulting from the analysis on the variables of set 3 is presented in table 48. Together the variables 'cigarette consumption', 'cigar consumption', 'person-hours' and 'use of ventilation provisions in the living room' explained 48 % of the variance in the RSP levels between homes. The sign of the regression coefficient of the latter variable was contrary to the expectations. For the data of Ede separately, the variable 'person-hours' was not selected in the equation, but reached border line significance. For the Rotterdam homes, the variable 'use of ventilation provisions' was not selected.

Instantaneous RSP measurements

Instantaneous RSP measurements were carried out in kitchen, living room and bedroom, irrespective of smoking of the occupants. In some homes the measurements were obtained during or just after smoking, while in other homes

no tobacco had been consumed on the day of the measurements. Smoking during or prior to the measurements was registered in the questionnaires.
The geometric mean and range of the instantaneous RSP concentrations in the 3 indoor locations are presented in table 49. Table 50 shows the geometric mean RSP levels in the living room, broken down by prior tobacco consumption. The information about prior smoking explained 53 % of the variance in the logarithm of the instantaneous RSP levels in the living room.

Pearson correlation coefficients were calculated between the logarithm of the instantaneous RSP concentrations in the 3 indoor locations, to establish the relation between simultaneous RSP levels in different locations of the home. During, or within 0.5 hour after smoking, the correlation of the RSP concentration in the living room with that in the kitchen and in the bedroom was 0.44, respectively 0.40 ($n=75$, $p < 0.001$). For all homes combined, these correlations were 0.57 and 0.64 ($n=187$, $p < 0.001$).

5.2. Repeated measurement program.

Weekly average RSP levels were determined during 16 weeks between July 1982 and March 1983 in 4 of the homes of the repeated measurement program. The mean and range of the weekly average RSP levels in the 4 homes are presented in table 51. The homes A and B had non-smoking occupants, home C and D had smoking occupants.
Participants were asked to report the tobacco consumption in the home during the measurements weeks. After a few weeks, the reported tobacco consumption in 1 of the smoker's homes was always the same, probably because it was felt that repeatedly keeping track of tobacco consumption was too trying. Therefore, it was not attempted to correlate weekly tobacco consumption with the weekly average RSP levels.
Figure 17 shows a plot of the weekly average RSP levels in the 4 homes versus time. During the measurements in week 43 and 47, a smoking guest stayed in home A. In these weeks, the RSP levels in home A were clearly higher than in the other weeks. In home B, the highest RSP level was found in week 35, when 15 cigarettes were smoked by guests.

An analysis of variance was carried out to calculate the reliability coefficient for a single week-long RSP measurement. Unlike the analysis for repeated NO_2 levels, the analysis was not carried out for different seasons, since the measurements only covered a period of half a year. Due to missing values, the RSP levels of 13 weeks remained for the analysis of variance. The reliability for a single week-long RSP measurement was 0.69,

indicating that about 30 % of the total observed variance could be considered as error variance and 70 % as true variance.

5.3. Summary and discussion.

In many homes, the observed weekly average RSP levels in the living room were considerably higher than the typical outdoor levels of particulate matter. On average, indoor RSP levels were about 55 - 60 $\mu g/m^3$, but a wide range was observed. Instantaneous RSP levels were highest during and just after smoking. Peak-concentrations up to 1000 $\mu g/m^3$ were observed.
Elevated weekly average and instantaneous RSP levels were clearly associated with the tobacco consumption in the homes. The association of the variables 'family size' and 'person-hours' with weekly average RSP levels suggested an additional influence of indoor activity on weekly average indoor RSP levels.
Weekly average RSP levels in homes of non-smokers were about 30 $\mu g/m^3$. The contribution of tobacco consumption to weekly average RSP levels in the living room, estimated from the regression equations with questionnaire information as independent variables, was 2 - 5 $\mu g/m^3$ per cigarette smoked per day and about 10 $\mu g/m^3$ per cigar per day.

The sign of the association between the variable 'use of ventilation provisions in the living room' and indoor RSP levels indicated that long use of ventilation provisions was associated with higher RSP levels. Following similar reasoning as for kitchen NO_2 levels, this might indicate that the perceived pollutant levels (for instance irritating properties of tobacco smoke) stimulate occupants to increase the ventilation. van Dongen (1984) observed that in homes of smokers the living room was ventilated twice as long as in identical homes of non-smokers. In the homes of the week-long measurement program, the correlation between tobacco consumption and use of ventilation provisions was low. In different homes of identical structure, pollutant levels caused by the smoking of a cigarette will generally show less variation than in homes with substantial differences in geometry like the homes in the week-long measurement program. It is therefore probably not tobacco consumption as such, which leads to increased use of ventilation provisions, but the actual pollutant levels from smoking. The positive association between use of ventilation provisions and indoor RSP levels, however, suggests that the increased ventilation is unsufficient to substantially reduce the indoor pollutant levels caused by smoking. In addition, there were indications that occupants often use the ventilation

provisions at the wrong moment, i.e. during the night (so that the air in the living room is 'fresh' the next morning), instead of during the pollutant production in the occupied room, when ventilation would be most effective.

A considerable variance was observed in the simultaneous RSP levels in different locations of the home. The instantaneous RSP measurements can be considered as repeated measurements of RSP at different locations, and the regression coefficient of 2 repeated measurements can be interpreted as a reliability coefficient. Thus, the correlation coefficient between simultaneous RSP levels indicate that, when instantaneous living room RSP levels are used to describe (exposure to) the RSP level in the home, 40 to 60 % of the variance in the RSP level can be considered as error variance.

Comparable or somewhat lower indoor RSP levels than those observed in the week-long measurement program, have been reported for homes in the USA. Moschandreas et al. (1978) measured RSP levels in homes and found 24-hour average levels of 5 - 260 $\mu g/m^3$ when smokers and/or small children were present, and 1 - 8 $\mu g/m^3$ in homes without smokers and small children. The association between the presence of small children and indoor RSP levels was considered to reflect the effect of indoor activity on (re)suspension of particulate matter.
Large numbers of indoor samples of RSP were taken in the Harvard Six Cities Study (Spengler et al. 1980, Dockery and Spengler 1981, Ju and Spengler 1981, Spengler et al. 1981). Annual mean RSP levels were generally lower than 60 $\mu g/m^3$. Indoor levels were usually higher indoors than outdoors; a clear relationship with tobacco consumption was observed. Dockery and Spengler (1981) reported a RSP concentration increase of about 1 $\mu g/m^3$ per cigarette smoked per day in not fully air conditioned homes.
Results of a Yougoslavian study showed indoor RSP levels between 131 and 204 $\mu g/m^3$ during the winter period, in an area where outdoor RSP levels ranged from 151 to 246 $\mu g/m^3$. Regression analysis suggested that a considerable part of the indoor RSP was (re)generated indoors (Fugas et al. 1982). Peak-concentrations of RSP in homes in the USA of several hundreds of $\mu g/m^3$ during smoking, were reported by Repace and Lowrey (1980).
In the Netherlands, monthly average RSP levels were determined in 84 homes near a secundary lead smelter (Diemel et al. 1981, Brunekreef and Boleij 1982). Monthly average RSP levels ranged from 20 to 570 $\mu g/m^3$, with a geometric mean of 120 $\mu g/m^3$. Levels in the first and second measurement period were highly correlated (r=0.79, n=76); with 2 repeated measurements, the correlation coefficient (r) can be interpreted as a reliability coefficient (RC)(cf. part 1, section 3.6). A clear relation was observed between indoor RSP levels and the number of smoking occupants.

In the homes of the repeated measurement program, the average indoor RSP levels in the 2 smoker's homes were consistently higher, on average by about 60 $\mu g/m^3$, than the levels in the non-smoker's homes.Highest RSP levels in the homes of non-smokers were observed during the stay of smoking guests. The reliability coefficient of a single week-long measurement of RSP was 0.69. Although this RC was calculated from data of only 4 homes, the reported high correlation between monthly average indoor RSP levels from 2 sampling periods (which can be interpreted as a reliability coefficient), indicates that also in other Dutch homes the indoor RSP levels are rather stable. The observed error variance in weekly average indoor RSP levels within homes of about 30 %, cannot be explained by questionnaire information about the average tobacco consumption. Regression models incorporating the actual tobacco consumption determined by the diaries, did not yield better results than those with questionnaire information. The accuracy of the RSP levels predicted with regression models was only about 30 % better than the accuracy of predictions without regression models.

The 24-hour average maximum values of the (proposed revision of) the USA primary standard for outdoor particulate matter were regularly exceeded in the homes of the week-long measurement program. Given the relative stability of indoor RSP levels observed in the repeated measurement program and reported by Brunekreef and Boleij (1982), it is likely that also the lower annual values of the standard will be frequently exceeded in Dutch homes.
Just as for NO_2, it is unclear to what extent indoor RSP levels may lead to adverse health effects. Most health effect studies have dealt with the risk of passive smoking and not with exposure to indoor RSP as such. In a recent review, Weiss et al. (1983) concluded that passive smoking leads to an increased frequency of acute and chronic respiratory symptoms in children. The results of different studies on the association between pulmonary function and passive smoking were considered inconclusive. Recent preliminary results of a Dutch study on the health effects of indoor exposure to NO_2 and passive smoking showed that several pulmonary function parameters were significantly associated with tobacco consumption in the homes (Brunekreef et al. 1985). Just as for NO_2 these preliminary results need further confirmation.
van Houdt et al. (1984) reported results of a study on the mutagenic activity of TSP, inside and outside Dutch homes(mutagenic activity refers to the ability to invoke changes in the genetic structure of living cells). Using the Ames-test(a test for the mutagenic effect on bacteria), they found that indoors the mutagenic activity(after metabolic activation) was higher than outdoors; direct mutagenic activity lower indoors than outdoors. Cytotoxic effects were only found indoors. Tobacco smoke was reported as the most important contaminant indoors for mutagenic activity.

CHAPTER 6. RESULTS; VOLATILE ORGANIC COMPOUNDS.

In chapter 6, the results of the week-long measurement and repeated measurement program will be presented. Week-long VOC measurements were carried out in 3 age-groups of homes: in post-war homes in Ede during the winter of 1981/82, and in the following winter in pre-war homes in the inner-city of Rotterdam and in less than 6 years old homes in Ede.
The repeated VOC measurements were obtained in the same 4 homes in which repeated RSP measurements were carried out. In addition repeated measurements of VOC were carried out in 11 newly built homes before and during the first 3 month of occupation. The results of the measurements in the newly built homes will be treated separately.

45 of the most abundant coelutent free volatile organic compounds in the boiling point range of 70 to 270 °C were determined quantitatively. 44 of these VOC were grouped in 5 classes for the purpose of data summarization. This classification was based on the chemical structure of the compounds (table 52). Results for separate compounds as well as for the 5 groups of VOC will be presented. Limonene was treated separately in the data analysis. For reasons which will be explained in the discussion, differences in VOC levels between the age-groups of homes were not tested on statistical significance.

6.1. Week-long measurement program.

Successful VOC measurements were obtained in 134 post-war homes in Ede, in 89 pre-war homes in Rotterdam and in 96 less than 6 years old homes in Ede. Table 53 shows the percentage of homes with VOC levels above detection limit, the median and maximum concentration of the 45 VOC in the 3 age-groups of homes and outdoors, and also the ratio of median indoor to median outdoor levels are given in table 53. Figure 18 shows the frequency distribution and the geometric mean and maximum concentration of the 5 groups of hydrocarbons.

VOC were detected in a vast majority of the homes. Compounds which were detected in only a minority of the homes were: dimethylcyclopentane isomers, i-propylbenzene, naphtalene, 1-methylnaphtalene and, with the exception of p-dichlorobenzene, the chlorinated hydrocarbons. Indoor levels of

most VOC had a wide range and were generally higher, sometimes orders of magnitude higher, than the outdoor levels. Especially the straight-chain hydrocarbons from C9 to C12 had indoor-outdoor concentration ratios higher than 10; limonene had an indoor-outdoor ratio of more than 80. An illustration of the differences between indoor and outdoor VOC levels is presented in figure 19, which shows the chromatographic patterns of an indoor and outdoor sample.

The measures of central tendency of the distribution of VOC levels had, in general, comparable values in the 3 age-groups of homes;the geometric mean of the total straight-chain hydrocarbon concentration, however, was about 50 % higher in the group of less than 6 years old homes than in the other age-groups. The median limonene concentration in the homes of less than 6 years old, was about 20 $\mu g/m^3$ higher than in the other age-groups. Concentrations above 100 $\mu g/m^3$ for 1 or more of the VOC were found in 14 % of the post-war homes, 17 % of the pre-war homes and in 29 % of the less than 6 years old homes.

Interdependencies among VOC levels.

From the chromatographic patterns it was clear that elevated indoor levels of several compounds were associated with each other; this was corroborated by the observation that for 17 compounds, maximum concentrations in the group of less than 6 year old homes were found in only 3 different homes. It was assumed that when several compounds share the same source, use of this source (when powerful enough) will lead to correlated VOC levels in the homes. A measure for the strength of such interdependencies among indoor levels is the squared multiple correlation (SMC) of the level of each compound with the levels of all the others. The SMC can take values between 0 (no association) and 1 (complete association).
SMC values were calculated separately for the 3 age-groups of homes, after logarithmic transformation of the data. Most compounds had SMC values of over 0.5 and often of 0.8 and higher, which means that a large fraction of the variance in the concentration of one compound could be explained by the levels of the other compounds. SMC values of less than 0.5 in one or more of the 3 age-groups of homes were found for n-hexadecane, limonene, naphtalene and 1-methylnaphtalene, tri- and tetrachloroethene, and momo-, di-, and trichlorobenzenes.

Factor analysis was used as an exploratory tool to delineate the underlying patterns of the observed high SMC values, by clustering the highly corre-

lated compounds in 1 or a few groups. (cf. part 1, section 3.6). Factor loadings, expressing the degree of association of a compound with a factor, were calculated separately for the 3 age-groups of homes by Maximum Likelyhood Factor Analysis (with oblique rotation), after rank-order transformation of the data. In this way, 2 clusters, consisting of more or less the same compounds in the 3 age-groups of homes, were tentatively identified. In table 54 these 2 clusters are visualized by indicating compounds with a factor loading of 0.5 or higher with a ' + '. (A factor loading of 0 indicates that a compound is not involved in the cluster, a value of 1 indicates that the compound is highly associated).

Compounds with factor loadings higher than 0.5 in at least 2 of the 3 age-groups of homes were:

Cluster 1 :n-hexane, n-heptane, 3-methylpentane, 2- and 3-methylhexane, cyclohexane, methylcyclohexane and dimethylcylopentanes
Cluster 2 :n-nonane, n-decane, n-undecane, xylenes, ethylbenzene, methylethylbenzene, 1,2,4- and 1,3,5-trimethylbenzenes, n-buthylbenzene and p-methyl-i-propylbenzene

The clusters resemble the VOC mixtures in common petroleum-based solvents with different boiling ranges.

Associations between VOC levels and the use of possible indoor sources of VOC.

The effect of the use of possible indoor VOC sources on the indoor VOC levels was tested in a statistical analysis with the logarithm of the 5 groups of VOC and limonene as dependent variables. Two additional dependent variables were created to represent the 2 clusters of VOC which were described in the previous section. These dependent variables were formed by summation of the standardized concentration scores (z-scores) of those VOC which formed a cluster. Standard scores of the VOC levels were used to make the contribution of the different compounds in a cluster independent of their concentration ranges. Thus, for each cluster, a value which summarizes and represents the level of 2 groups of several related VOC was assigned to each home.
The information on the use of possible indoor sources of VOC during the measurement period was used as independent variables (cf. part 2, section 2.2.6). Although the measured VOC cannot be considered dominant constituents, nor typical indicators of tobacco smoke, most of the VOC have been identified in tobacco smoke. Therefore the presence of smokers in the home

was used as an additional independent variable.

After initial bivariate analysis, multiple regression analysis was used to adjust for possible interdependencies among the explanatory variables. A significance level of 0.10 was used as selection criterium.
All independent variables were dichotomous; all dependent variables were logarithmically transformed (cluster scores were raised by 10 to avoid transformation of zero or negative values).
The results of the regression analysis, which was carried out on the data of the 3 age-groups of homes separately as well as combined, are summarized in table 55.
As could be expected from the observed wide range in the VOC levels and the crude classification of source use, only a minor part of the variance in the dependent variables could be explained by the selected independent variables. Only for the straight-chain and aromatic hydrocarbons and for cluster 2, the R^2 of the regression equations exceeded 0.10, when the data of all homes were combined.
Table 56 illustrates the associations of solvent use and presence of smokers with the levels of straight-chain and aromatic hydrocarbons.

6.2. Repeated measurement program.

The repeated measurements of VOC resulted in 14 valid observations for each of the 4 homes. The concentrations of the di- and trichlorobenzenes of this series of samples could not be quantified reliably; the data for these compounds were discarded.
The median and maximum VOC concentration in each home and the reliability coefficient for the repeated measurements are given in table 57. The VOC levels in the 4 homes had about the same range as was found in the week-long measurement program. The median concentrations of the higher straight-chain hydrocarbons in home A, however, were high in comparison to the median concentrations found in Ede and Rotterdam. Several compounds had a wide range in some homes, but not in others; especially home D had a wide concentration range for many of the compounds.
The RC of the compounds in the 4 homes varied between 0.01 and 0.96. Most of the higher RC were found in the group of straight-chain hydrocarbons. This is largely due to the relative stable and high levels of these compounds in home A. On the other hand, many of the lower RC were dominated by the wide range in the VOC levels in home D. In this home, many compounds had there maximum concentration in 1 single week, in which the concentrations of at least 16 VOC were several times higher than in other weeks;

among these were 10 of the compounds from which the clusters in table 54 were built up.

The differences in concentration levels both between homes and within homes are illustrated in the figures 20a,b and c, which show the levels of n-undecane, limonene and the total hydrocarbon concentration in the 4 homes versus time. n-Undecane is an example of a compound with relatively stable indoor levels in these homes. Limonene, on the other hand, is an example of a compound with unstable indoor levels. Figure 20c shows that week 47 in home D is an aberration of all other weeks, with a total VOC concentration of 2283 $\mu g/m^3$ (the mean of the total VOC level in home D was 337 $\mu g/m^3$). This very high level was probably due to hobby and home craft activities in that week. The limonene level in the 4 homes and the total VOC level in home B, C and D seem lower in August and September, than in the rest of the measurement period. This might be the result of higher home ventilation in these month with relatively mild weather. The measurement period of half a year, however, is considered too short to discover seasonal patterns.

6.3. Measurements in newly built homes.

Successful repeated measurements of VOC levels in newly built homes under unoccupied and occupied conditions were carried out in 11 homes. Figure 21 shows the minimum, geometric mean and maximum concentration in these homes at different points in time for the 5 groups of hydrocarbons. The letters on the horizontal axes in this figure indicate the different phases before and during occupation, when the measurements were carried out:

A :empty homes, before occupation
B :just after floor- and wall covering was carried out
C :just after the homes were furnished
D, E and F :occupied homes, respectively during the first week of occupation, after 1 and after 3 months of occupation

In 2 of the homes, the floor- and wall covering and furnishing were carried out simultaneously; the results of the measurements just after these activities were classified under phase B.

The median VOC levels found in the week-long measurement program were used as a reference level for 'typical' outdoor and 'typical' indoor concentrations; these reference levels are indicated in figure 21 by the dotted line.

The straight-chain and aromatic hydrocarbons, as well as the branched-chain and alicyclic hydrocarbons had similar concentration patterns. In the empty homes (phase A), the straight-chain and aromatic VOC were generally higher than the indoor reference levels, but well within the range found in the week-long measurement program, where maximum concentrations were observed of over 1000 $\mu g/m^3$.The branched-chain and alicyclic compounts had concentrations at, or below, the outdoor levels at phase A.
The floor- and wall covering activities (phase B) led to elevated levels of the non-chlorinated compounds, with large differences between homes.
At phace C, the straight-chain hydrocarbon concentrations already dropped to about the same level as was found in the empty homes. The branched-chain and alicyclic hydrocarbon levels dropped on average to the reference level from the week-long measurement program. Aromatic hydrocarbon levels were lower than at phase B, but remained higher than in the empty homes.
In the occupied homes, again a wide range in the levels of the non-chlorinated compounds was observed. The straight-chain and especially the aromatic compounds had lower levels than at phase C, but on average remained above the reference levels for indoor concentrations of 50 $\mu g/m^3$ and 75 $\mu g/m^3$, respectively. Branched-chain and alicyclic compound levels in the occupied homes fluctuated around the level at phase C, which is comparable to the reference levels from the week-long measurement program.
Several compounds were only observed at elevated levels in the occupied homes and not in the empty unoccupied homes. These were : n-tetradecane, n-pentadecane, n-hexdecane, limonene, naphtalene and 1-methylnaphtalene.

The levels of the chlorinated hydrocarbons did not show such large fluctuations as the non-chlorinated compounds. In general, the chlorinated hydrocarbons levels were higher than those found in the week-long measurement program. Although the sum of the concentrations of the chlorinated hydrocarbons was stable in the 6 phases, the concentration of the individual compounds was not. In the phases A, B and C the dominant chlorinated compounds were tetrachloromethane and chlorobenzene, while in the occupied homes at phase D, E and F, p-dichlorobenzene was most abundant.Tetrachloroethene and p-dichlorobenzene were not observed in the occupied homes.

6.4. Summary and discussion.

In occupied homes, virtually all VOC had higher indoor than outdoor levels. Most of the measured VOC have been reported as indoor pollutants in schools and offices and have been identified in the head space of building materials (Jarke 1979, Berglung et al. 1982, Miksch et al. 1982, Mølhave 1982,

Kuwata et al. 1983, Girman et al. 1984, Montheith et al. 1984).
For several VOC, levels comparable to those observed in this study, were measured in 15 flats in Berlin (Seifert and Abraham 1982) and in 14 homes in Northern Italy (De Bortoli et al. 1984). Toluene and xylene levels in the homes of the week-long measurement program were lower than those reported for bedrooms in 39 Danish dwellings (Mølhave and Møller 1979). The comparisons should be interpreted with some care because of the differences in measurement methodology and strategy.

It was felt that statistical tests of differences in indoor VOC levels between age-groups of homes would not add much to the understanding of indoor VOC.
First, the age-groups were selected to be different in age, location and building type, but differences in a.o. family size, marital status and age of the respondents were also observed. Even if differences in the VOC levels between age-groups of homes would be considered significant, this would not provide a clue to what caused these differences, since the nature of the indoor sources is largely unknown at this stage.
Second, there is the question of how to test.Should the age-groups be compared on differences in median or extreme values? Should all the observed differences be in the same direction, i.e. higher in one age-group than in the others?. The choice of the statistical instrument, the data transformation procedure and corrections for multiple comparisons would be largely arbitrary and these choices would affect the results of the tests.
Apart from chance, the differences between the groups of homes might result from lower ventilation, more frequent use of sources or a higher emission from relatively new building materials in the youngest age-group of homes.

The clustering of several of the solvent type compounds suggests the existence of some common sources for these hydrocarbons. It is, however, difficult to distinguish between building materials and consumer products as a source. For instance, mixtures of compounds similar to the ones in the 2 clusters have been identified as solvent in building materials (Mølhave 1982, Girman et al. 1984), but were also found in the head space of consumer products like shoepolish and furniture polish. Toluene, widely used as a solvent in building materials, is also constantly emitted indoors from the ink of newspapers, magazines and printed publicity material. Toluene emission rates of 150 to 400 $\mu g.kg^{-1}.hr^{-1}$ during the first 3 days have been established for fresh magazines in the laboratory. It is interesting to note that toluene had factor loadings of about 0.4 on both clusters in the factor analysis, which might indicate some degree of involvement in the solvent type of VOC mixtures, but also with other sources not related to other VOC.

From the measurements in newly built homes, it appears that during the first 3 months of occupation the straight-chain and aromatic hydrocarbon levels are generally higher than in older homes; the application of floor- and wallcovering materials gave transient concentration peaks for the non-chlorinated compounds.
The maximum concentrations found in the week-long measurement program were much higher than the concentrations in the new empty homes and than the average levels during the first 3 months of occupation. This suggests that these maxima were caused by incidental occupant activities and not by a constant high emission from new building materials. The low reliability coefficients of many VOC levels in the repeated measurement program support this view. The high and stable levels of several higher straight-chain hydrocarbons in one of the homes of the repeated measurement program, however, indicate an important constant source for these VOC.

It should be noted that the 45 measured VOC are not the only indoor VOC, but the ones which could be quantified reliably on a routine base. Numerous other compounds were detected in the gaschromatograms, but were not quantified. It should also be stressed that the observations about the relations among indoor VOC levels and about the associations between indoor VOC levels and possible sources of VOC have a tentative character and need further confirmation. There was, however, a general consistency in the results of the 3 age-groups of homes and the results of the different measurement programs were in support of each other. The results of the study indicate that most of the measured VOC have both consumer products and building materials as indoor sources. It is thus unlikely that a single week-long measurement of indoor VOC will provide a good impression of typical VOC levels in a particular home. Without further knowledge about the nature of indoor VOC sources and their patterns of use, it is unclear how often and over which time period the air in homes should be sampled.

Most of the research on the effects of VOC has been carried out on isolated compounds in (animal) experiments or in occupational settings. Little is known about the effects on the general population of long-term exposure to a combination of low level VOC. It is therefore difficult to evaluate the observed VOC levels from a health perspective. The measured indoor levels were well below occupational standards and known effect levels, even when peak levels several times higher than the weekly average levels are considered. Occupational standards, however, are set to limit the exposure of healthy adults to a single compound during 40 hours per week. These standards are therefore unsuitable to evaluate the health risk for the general population of exposure to VOC in the home environment.
Mølhave and co-workers studied human reactions during controlled exposure

to low levels of mixtures of VOC (0, 5 and 25 mg/m^3 total VOC level) known as normal indoor pollutants (Bach et al. 1984, Møhave et al. 1984). The authors reported that performance on a memory impairment test and scores on several aspects of perceived air quality (administered by questionnaires), significantly decreased during exposure.

It has been suggested that indoor VOC levels may be part of the explanation of some symptoms of the 'sick building syndrom' (Mølhave 1982, Bach et al. 1984), but there is little practical evidence to support or refute this view.

CHAPTER 7. GENERAL DISCUSSION.

In the previous chapters, the observed indoor pollutant levels have been discussed separately. In this final chapter some more general aspects of the indoor air pollution problem will be discussed.

7.1. Study design and interpretation of the results.

For logistic reasons it was decided to determine the different pollutants under study as much as possible in the same homes. This approach had the obvious advantage of limiting the efforts of selecting and visiting homes. Furthermore, information from inspection lists, questionnaires and diaries could be used to explain the variance in indoor levels of several pollutants. A drawback of the approach was that the sampling period in the week-long measurement program was the same for all pollutants which were measured simultaneously. The choice of the week-long sampling period was based on the assumption that many occupant activities like cooking, smoking and ventilation are carried out in a daily routine. The diary information, observations of the real-time monitoring program and studies on time budgets (Knulst and Schoonderwoerd 1983), use of gas appliances (Dijkhof and Ogink 1978) and ventilation habits (van Dongen 1984) corroborated this assumption. Furthermore, the repeated measurements of NO_2 and RSP indicated that the variance in the levels of these pollutants within homes is relatively small as compared to the variance between different homes. One or a few repeated week-long measurements are therefore sufficient to establish differences in weekly average NO_2 and RSP levels between homes. For CO and VOC, the week-long measurement period was probably not optimal. Weekly average CO levels were low and differences between homes were small, although substantial differences in CO peak-concentrations were observed in the real-time monitoring program. For VOC, it is difficult to select the appropriate sampling period and frequency, since the nature of the indoor VOC sources is largely unknown.

There was a general consistency in the results of the 3 main measurement programs and the related studies. The results of the regression analysis showed that the relations between indoor sources and pollutant levels, observed in the real-time monitoring program, and in the case of CO ,under standardized conditions, are not restricted to a limited number of selected

homes, or unrealistic standardized conditions. These relations could also be observed in large groups of homes under normal living conditions.
The similarity between the observed NO_2 levels in the 275 homes of the week-long measurement program and those observed in another 700 homes in related studies indicate that, although the homes of the week-long measurement program cannot be considered to be a representative sample of the housing stock, they do represent large numbers of Dutch homes.
The assumption that inner-city pre-war homes might deviate unfavorably from other groups of homes with regard to indoor pollutant levels, was only partly correct. Only the indoor NO_2 levels in the pre-war homes in Rotterdam were higher than those in Ede; after adjustment for independent variables, NO_2 levels in kitchens and living rooms were not significantly different between towns, but bedroom levels remained higher in Rotterdam than in Ede.

The repeated measurement program indicated that about 15 - 20 % of the variance in indoor NO_2 levels was within-home or error variance. For RSP, the within-home variance was about 30 % in the 4 homes. The major part of the remaining (between-home) variance for NO_2 and RSP could be explained by the multiple regression models. In the course of the regression analysis, special care was taken to detect possible collinearity among the independent variables. (Multi-)collinearity refers to the situation in which independent variables are highly interrelated; in that case, a regression coefficient cannot be considered as the marginal effect of an independent variable on the dependent variable. For the variables used in the regression analysis, there were no indications for the existence of a high degree of collinearity. Nonetheless, it should be kept in mind in the interpretation of regression coefficients, that a variable like 'use of gas cooker' in the regression equation for kitchen NO_2 levels, may incorporate some of the effects of 'use of unvented geiser', 'use of pilot light of gas cooker' and 'use of pilot light of geiser' on kitchen NO_2 levels. Likewise, the variable 'number of smokers' may not only represent the effect of tobacco consumption as such on indoor RSP levels, but also some effect of indoor activity, which in other equations was represented by variables 'family size' and 'person-hours'.

Although it is difficult to evaluate the accuracy of the diary information, the diaries were considered a useful tool to assess occupant activities. After discarding the diaries which were judged as inaccurate during the hand coding procedure, no clear discrepancies were observed in checks of diary information against information about the position of ventilation provisions and interior doors during home visits. Also comparisons of the results of diaries judged as 'sufficient' with those of diaries judged as

'good', and comparisons (as far as possible) of diary results with questionnaire information, agreed well.
The most prominent effect of using diary information in the regression analysis was the reduction of the number of homes with a complete set of data. Regression models with diary information could not explain more of the variance in indoor pollutant levels than models with less sophisticated data as independent variables. Yet the diaries yielded some interesting observations. The wide range in the diary results clearly demonstrated that defining 'typical' occupant activities (for cooking or ventilation) is an undue simplification which underrates the activities of a considerable part of the population. Furthermore, the observed positive associations between use of ventilation provisions and NO_2 levels in the kitchen and RSP levels in the living room, suggest that indoor pollutant levels as experienced by the occupants (water vapor from gas appliances, irritating agents from tobacco smoke) may act as an incentive to increase the use of ventilation provisions. However, the resulting additional ventilation may not be enough to reduce the indoor pollutant levels, possibly because the ventilation provisions are used at the wrong moment.

During the field work of the study some general and qualitative observations, which have bearing on the problem of indoor air pollution, were made. These observations are rather trivial, but in the literature and discussions on indoor air quality there is often little recognition for such observations, possibly because of the general qualitative nature.

-Occupants exhibit a distinct and consistent ventilation behaviour, but most occupants do not often dwell upon the why and how of their ventilation behaviour. Respondents often felt it difficult to answer questions on ventilation habits, because they did not realize at first what these habits were.

-Occupants make many changes in their home. Interior walls and doors are sometimes removed, ventilation ducts are plugged or inappropriatly connected to a range hood. The location of ventilation provisions, in particular vent lights, and the desired location of objects like washing machines in the kitchen and plants in windowsills prevent occupants from using their ventilation provisions in the way the home designer might have intended.

-About 20 % of the respondents in Ede and Rotterdam felt that the ventilation provisions in kitchen and living room were inadequate to maintain the desired ventilation. For bedrooms this was 1 % respectively ± 2 % in Ede and Rotterdam. Even in the group of homes of less than 6 years old, probably all built according to the present building standards, 20 % of the respondents judged the ventilation provisions in the living room as inadequate (for kitchens and bedrooms no information is available).

7.2. Indoor pollutant levels and outdoor air quality standards.

Air quality standards for outdoor air pollutant levels provide a framework for a first evaluation of indoor air quality. In contrast with occupational standards, these outdoor air quality standards generally aim at protecting the general population, including sensitive individuals. There are, however, some difficulties in comparing indoor pollutant levels to outdoor air quality standards.

First, these air quality standards are often partly based on epidemiological studies, which attempted to relate health effects to outdoor pollutant levels. The results of this and other studies on indoor air pollution, indicate that exposure to indoor pollution may have introduced bias, which can lead to under- or overestimation of the health effects. This bias may have influenced the setting of air quality standards. Thus, there is a paradox in comparing indoor levels with outdoor air quality standards.

Second, the conditions of exposure to air pollutants in homes may be different from the conditions for which the standards were developed. This, for instance, may be the case for CO, where the indoor exposure is probably associated with exposure to elevated CO_2 levels, which may increase the respiration rate, and also with exposure to NO, which binds hemoglobin to produce methemoglobin. It has been suggested that many of the adverse health effects reported in the past for CO alone, may be related to the combined action of COHb and methemoglobin (National Research Council 1981). Furthermore, the indoor particulate matter in homes of smokers will be of a different nature than outdoor particulate matter and elevated RSP levels indoors will be associated with elevated levels of other constituents of tobacco smoke.

For a further evaluation of the health risks of air pollutants, studies are needed which incorporate indoor as well as outdoor exposure to air pollution.

7.3. Indoor air pollution and environmental epidemiology.

The results of this study indicate that in the Netherlands, air pollution in homes will give a substantial, if not dominant contribution to personal exposure of the general population to air pollution for the pollutants which were studied. For NO_2 , this has recently been confirmed in 2 personal exposure studies (Hoek et al. 1984, Noy et al. 1985).

In the past, with its more severe local outdoor air pollution episodes than nowadays, crude exposure estimates, based on regional differences in outdoor air pollution levels, have been used with success in epidemiological studies. Furthermore, the episodic character of outdoor air pollution made

it possible to study shifts in daily mortality and morbidity indices (Biersteker 1966). Because outdoor levels of general air pollutants have declined over the last few decades, the importance of exposure to indoor air pollution has grown and the nature of exposure has changed. Exposure to air pollution has no longer a regional but an individual character; the episodic nature of exposure has changed into a more chronic one. This calls for a more individual approach of exposure in epidemiological studies on the health effects of air pollution.

Recalling the definition of microenvironments as locations with a homogeneous pollutant concentration in space and time (cf. part 1, section 1.2), it is clear that Dutch homes cannot be considered as microenvironments, since considerable differences between pollutant levels in different locations of the home have been observed. Evenmore, by this definition, a single room is not a microenvironment because of the short-term fluctuations in pollutant levels. The personal exposure of an individual to indoor air pollution will therefore largely depend on at which location in the home he or she is during the activities which generate indoor air pollution. However, being in the living room during pollutant production in the kitchen may not always protect the occupant from exposure to peak-concentrations, since the communication of air between different locations in the home may vary from day to day. Defining the home as a microenvironment with respect to exposure to air pollution may provide the false notion that this exposure can adequately be described by measuring the pollutant levels at one location in the home.

In environmental epidemiological studies, multiple regression analysis is an often used technique to relate health parameters of participants to the pollutant exposure of the participants. One of the basic assumptions in regression analysis is that the independent variables, i.e. the exposure estimates, are free of random error, a condition which is seldom met. (Note that the following reasoning similarly applies to the earlier discussed regression analysis which was used to explain indoor pollutant levels).
Random error in the independent variable not only reduces the statistical power of the test (i.e. reduces the chance to find a significant association if one exists), it also introduces bias in the estimates of the regression coefficients and thus in the estimated exposure-response relation (Cochran 1968, 1970). The random error in an exposure variable can be caused by errors in sampling and analytical techniques of the measurement method, but also by variability of the pollutant levels in time and place. The definition of the error in the exposure estimate must be viewed from the perspective of the health effects under study. For instance, for the development of respiratory disease in pre-school children, the relevant

exposure to NO_2 and passive smoking covers a period of several years. The reliability of a single exposure measurement is thus determined by the variability of the exposure over this time period. The reliability can be assessed by repeated measurements of the exposure, analogous to the repeated concentration measurements discussed earlier.
In a bivariate regression model, the bias in the regression coefficient is proportional to the reliability coefficient of the exposure variable and therefore toward zero; the bias is independent of the size of the study population. In a multivariate regression model, the bias in the regression coefficients can be toward as well as from zero, and the reliability of one independent variable may affect the regression coefficient of another independent variable (Cochran 1968).
Considering that exposure to peak-concentrations of NO_2, and to RSP/passive smoking over a period of years may be the relevant exposure for the development of respiratory disease and pulmonary function decline, it is obvious from this and related studies on indoor air pollution, that errors in the presently available exposure estimates (i.e. presence of gas appliances, smokers, or a single week-long indoor measurement) may affect the results of health effect studies in this field in an unknown manner. Since bias in regression coefficients cannot be remedied by increasing the size of the study population, more reliable exposure estimates are badly needed.

7.4. Indoor air pollution and public policy.

In Dutch homes, the indoor levels of the pollutants under study were generally higher than the outdoor pollutant levels. A first evaluation of the observed indoor levels against outdoor air quality standards learned that (keeping the earlier mentioned restictions in mind) the levels of CO, NO_2 and RSP cannot be considered safe for the general population. Although the results of the few studies on health effects of indoor NO_2 and RSP/passive smoking are inclonclusive, probably not in the least because of the poor reliability of the available exposure estimates, several studies indicate that the observed pollutant levels such as found in Dutch homes, may represent a health risk.

Recently, a Working Committee of the Dutch Health Council has published an advise on minimum standards for home ventilation from a public health perspective (Dutch Health Council 1984, van der Kolk 1984). The Committee concluded that indoor pollutant sources could be classified as unavoidable, i.e. occupants themselves, and avoidable sources, i.e. all other sources. The present ventilation standard, providing for a minimum fresh air supply

of 25 m^3 per hour per occupant, was considered adequate to guarantee proper indoor air quality in the presence of the unavoidable sources. The Committee was not in favor of the promulgation of indoor air quality standards, because of the impractibility of control of indoor standards.
In the view of the Committee, all other sources than the occupants should be avoided or the source strength should be reduced as much as possible. Thus, improvement of indoor air quality should start at the source; the Committee proposed the implementation of product standards to limit the source strength of avoidable sources. In the setting of these product standards, a minimum fresh air supply of 5 m^3 per hour per occupant should be assumed, because many occupants make little use of their ventilation provisions and will not maintain a minimum fresh air supply of 25 m^3 per hour per occupant.
The responsibilities for proper indoor air quality are shared by government, home designers and -builders, gasindustry and -companies, product manufacturers, home owners and by the occupants themselves. However, the responsibilities are ill-defined and not generally recognized. The major responsibilities may be different for the different pollutants under study. For localized sources like gas appliances, local exhaust systems like range hoods and vents can be applied, but in the existing housing stock this may be difficult to achieve. When old gas appliances need replacement, electric appliances might be an alternative, when local exhaust systems cannot be applied. Burners with low CO and NO_2 emissions could also improve indoor air quality. For RSP (and other pollutants from tobacco smoke) the responsibility of the occupants is quite obvious. In the case of CO, occupants also have a responsibility for the proper maintenance and use of gas appliances. For VOC the situation is rather complicated. First, it is difficult to assess the health risks of the simultaneous exposure to a combination of low-level VOC with the present knowledge. Second, VOC are probably produced by building materials as well as consumer products, which makes it difficult to set standards for the different types of sources, when such would be judged necessary.

At present, lacking clear health criteria for the evaluation of indoor air pollution, a sensible and pragmatic approach would be to replace potentially harmful products and appliances by not or less harmful ones (Andersen, 1982, Dutch Health Council 1984). Thus, the old adages of environmental policy, i.e. the principle of 'the best practical means' and the 'stand-still' principle may prove valuable again in the evaluation of the eligibility of products and appliances for use in homes. For instance, one could reason that, although there is no scientific proof (yet?) for ill-health effects, unvented kerosene heaters are unfit for permanent use in Dutch homes, because these appliances would further elevate the already high

indoor NO_2 levels (violation of the stand-still principle) and other space heaters, which do not affect indoor air quality, are available (principle of best practical means).

As was and still is the case in environmental policy, policies to improve indoor air quality are likely to meet opposition, because they may bring about costs for the occupants and home owners and it may threaten the interests of product manufacturers and -suppliers. A more general awareness of the complex indoor air quality problem seems a prerequisite condition for the different groups in society to recognize their responsibilities.

SUMMARY.

This thesis deals with air pollution in Dutch homes from a public health perspective. It consists of two parts. Part 1 describes the history and background of indoor air pollution research and outlines the design of this study. In part 2, the results of the study are presented and discussed.

Part 1.

Chapter 1 presents the reasons for concern about indoor air quality: the campaigns to reduce domestic energy consumption, initiated after the "oil crisis" of 1973, the introduction of new materials and chemicals in the home, and, in addition, the time activity patterns of people in western industrialized societies. The concept of integrated personal exposure, illustrating the importance of the indoor environment for exposure to air pollution, is introduced. The background of the study is presented and the study objective is stated: characterization of the typical pollutant levels in Dutch homes in relation to properties of the home and occupants, from a perspective of integrated personal exposure and public health.
The pollutants under study, i.e. carbon monoxide (CO), nitrogen dioxide (NO_2), respirable suspended particles (RSP) and volatile organic compounds (VOC), are described briefly.
Chapter 2 gives an overview of indoor air pollution research methodology. Typical for the research in this field is that it originated from different disciplines and is marked by the perspectives of those disciplines. Three major research approaches are recognized and described: the mechanistic, case study and survey approach. The current state of knowledge on indoor air quality in general and, more specifically, for the situation in the Netherlands is described. It is concluded that the current state of knowledge, in particular for the situation in the Netherlands, is limited and of a fragmentary nature, but it justifies the concern about indoor air quality in Dutch homes.
Chapter 3 describes the study design and outlines the 3 main measurement programs, which together form the body of the study; the study design is summarized in the table on the following page:

Summarized study design

pollutants	measurement programs: real-time	week-long	repeated
CO	Wageningen - 12 homes - winter 82/83, 83/84 - k, l, b*	Ede - 174 post-war homes - winter 81/82 - k, l* Rotterdam - 102 pre-war inner-city homes - winter 82/83 - k, l*	
NO_2	Wageningen - 12 homes - winter 82/83, 83/84 - k, l, b*	Ede - 174 post-war homes - winter 81/82 - k, l, b* Rotterdam - 102 pre-war inner-city homes - winter 82/83 - k, l, b*	Wageningen and environs - 15 homes - March 82-Febr.83 - k, l, b*
RSP		Ede - 174 post-war homes - winter 81/82 - l* Rotterdam - 102 pre-war inner-city homes - winter 82/83 - l*	Wageningen and environs - 4 homes: 2 non-smokers 2 smokers - July 82-Febr.83 - l*
VOC		Ede - 174 post-war homes - winter 81/82 - l* Rotterdam - 102 pre-war inner-city homes - winter 82/83 - l* Ede - 103 homes ≤ 6 years old - winter 82/83 - l*	Wageningen and environs - 4 homes: 2 non-smokers 2 smokers - July 82-Febr.83 - l* Ede - 11 newly built homes - summer and autumn 83 - l*

* k = kitchen; l = living room; b = bedroom

Instrumentation, selection of the homes which were studied, field operation and data handling and analysis are described in chapter 3.

The objective of the real-time monitoring program was the determination of transient concentration peaks of CO and NO_2, the dispersion of pollutants through the home and day to day variation in pollutant levels. To determine kitchen ventilation, tracer gas experiments were carried out in this measurement program, using sulphurhexafluoride (SF_6); the use of unvented gas appliances was monitored with thermo couples.
The objective of the week-long measurement program was the determination of the distribution of CO, NO_2, RSP and VOC levels in a wide range of homes. In addition to the pollutant measurement, tracer gas experiments in kitchens were incorporated in the week-long measurement program. Additional information about the homes and about occupant activities was gathered with inspection lists, questionnaires and diaries.
The objective of the repeated measurement program was the determination of the variability of pollutant levels within and between seasons.

Part 2.

Chapter 1 outlines the organization of part 2.
Chapter 2 presents general characteristics of the homes which were studied. As far as appropiate, this chapter describes the use of indoor pollutants sources, the use of ventilation provisions and interior doors, and the results of tracer gas experiments. Emphasis is put on the presentation of those features of the homes and occupant activities that are used in the subsequent chapters in the data analysis to explain the variance in the observed pollutant levels.
For the 12 homes of the real-time monitoring program, examples are presented of data plots of the use of gas appliances and of the results of tracer gas experiments. In these homes, the cooking range was used 2 to 6 times per day; the geiser, when present, was used 10 to 35 times per day. Within homes, the use of gas appliances was more or less consistent from day to day, but large differences in appliance use were observed between homes. The results of the tracer gas experiments indicated inhomogeneous mixing of kitchen air. Large differences were found in the 4-hour average values of kitchen air-change rate and tracer gas transfer index within homes as well as between different homes.
The positive response in the week-long measurement program was satisfactory; over 50 % of the occupants which could be contacted agreed to participate in the study. However, the contact rate in the inner-city homes of

Rotterdam was low. Households with all members working full-time were probably underrepresented. Comparison of the distribution of characteristics of the homes of the week-long measurement program, like ownership, building type, cooking fuel, type of water and space heater with data from various sources revealed no clear deviations from this reference material. The distribution of general characteristics of the homes differed markedly in the 3 age-groups of homes. Wide ranges were observed in the use of gas appliances, ventilation provisions and interior doors, tobacco consumption, and also in the results of the tracer gas experiments. A considerable part of the respondents appeared not to use their ventilation provisions in the living room during the heating season.

Chapter 3 deals with the results of the CO measurements: the real-time monitoring program, the week-long measurement program, and two related studies.

For the homes of the real-time monitoring program, first some qualitative observations are presented, based on data plots. In these homes, the overall mean CO levels indoors were generally low, 0-3 mg/m^3 higher than outdoors, but short-term peak concentrations of several tens of mg/m^3, associated with the use of unvented gas appliances, were observed. CO concentration patterns were generally smoother in living room and bedroom than in the kitchen. The CO concentration patterns suggested a moderate effect of tobacco smoking on indoor CO levels of a few mg/m^3. If any, the margin between the air quality standard for outdoor CO (proposed by the Dutch Health Council) and the observed CO levels in the homes of the real-time monitoring program was small.

Weekly average CO levels in the homes of the week-long measurement program were in the same range as the overall mean CO levels found in the homes of the real-time monitoring program. Multiple regression analysis with the results of the week-long measurement program indicated that elevated CO levels in kitchens were associated with the presence of unvented geisers, burner type of the geiser, duration of use of the gas cooker and tobacco consumption. Living room CO levels were positively associated with kitchen CO levels and tobacco consumption, and negatively with the number of doors between kitchen and living room. In the regression analysis only a minor portion of the variance in CO levels in kitchens and living rooms could be explained. The observed effect of geiser and geiser burner type in the homes of the week-long measurement program was in good agreement with the results of a related study on CO production of geisers under standardized conditions.

The results of a related study on COHb levels indicated that under normal living conditions, the frequency of occurence of biological significant increases in COHb levels due to the use of geisers is less than the fre-

quency of geisers with a high CO production in the general housing stock.

Chapter 4 presents the results of the NO_2 measurements in the real-time monitoring program, the week-long and the repeated measurement program, and in related studies.
After the presentation of some qualitative observations for the homes of the real-time monitoring program, based on data plots, the 1-minute, 1-hour, 24-hour average and overall mean NO_2 concentration in these homes are presented. In general, indoor NO_2 levels were higher than outdoor levels. In several homes, 1-hour average peak concentrations of NO_2 of several hundreds of $\mu g/m^3$ were observed in all 3 locations.
On average, the ratio of the maximum 1-hour average concentration to the overall mean NO_2 concentration was 6 in the kitchen and living room and 4 in the bedroom. A considerable day to day variation in daily maximum 1-hour average NO_2 levels and in 24-hour average NO_2 levels was observed within homes. On some days in some of the homes, living room and bedroom NO_2 peak levels were just as high as the simultaneous kitchen levels, while on other days living room and bedroom levels were hardly affected by kitchen NO_2 levels.
In the homes of the week-long measurement program, indoor NO_2 levels also frequently exceeded outdoor levels. The observed levels were in good agreement with the levels found in 700 other Dutch homes in 4 related studies on indoor NO_2. In the inner-city homes of Rotterdam, NO_2 levels were generally higher than the levels in homes in suburban and rural areas. With multiple regression analysis, about 50 to 70 % of the variance in indoor NO_2 levels between the homes of the week-long measurement program could be explained. Presence of unvented geisers, presence and duration of use of gas cookers, outdoor NO_2 level, type of space heating and use of ventilation provisions in the kitchen were positively associated with kitchen NO_2 levels. Kitchen volume was negatively associated with kitchen NO_2 levels. Although no direct association was observed, the data analysis suggested that also the duration of use of the geiser was positively associated with kitchen NO_2 levels. Only for the data of Ede, a potential benificial effect of the use of the range hood on kitchen NO_2 levels was observed. In two of the related studies, similar associations between kitchen NO_2 levels and range hood were found. It is unclear whether the association between the variable 'type of space heating' and kitchen NO_2 levels means that local gas heaters sometimes act as direct sources of indoor NO_2 , or that this variable is a proxy for some other feature of the home. The positive association between the use of ventilation provisions in the kitchen and kitchen NO_2 levels might indicate that occupants increase the ventilation, when they perceive the indoor pollutant levels as high.
In the living room of the homes of the week-long measurement program, 60 %

of the variance in NO_2 levels could be explained by NO_2 levels in the kitchen, the number of doors between kitchen and living room and the type of space heating. When the variable 'tracer gas transfer index in the living room' was added to the regression model, 73 % of the variance in living room NO_2 levels could be explained.
About 50 % of the variance in bedroom NO_2 levels could be explained with the NO_2 levels in the other locations and outdoors, and the number of doors between kitchen and bedroom. After adjustment for these variables, bedroom NO_2 levels were significantly higher in Rotterdam than in Ede.
In the homes of the repeated measurement program, the variability of weekly average NO_2 levels in the 3 indoor locations was small during winter, spring and autumn, but larger during summer. In the kitchens of these homes, NO_2 levels were highest during winter and spring, intermediate in autumn and lowest in summer. In the other locations, no clear seasonal pattern was observed. The results of the repeated measurement program indicate that during the greater part of the year, indoor NO_2 levels were consistently different in different homes; these differences can be detected with one or a few measurements during the heating season.
The results of the real-time monitoring program as well as of the week-long measurement program indicated that the proposed air quality standard for outdoor NO_2 is frequently exceeded by kitchen NO_2 levels and regularly by living room and bedroom NO_2 levels.

Chapter 5 presents the results of the RSP measurements in the week-long and repeated measurement program. In addition to the weekly average RSP measurements, instantaneous RSP measurements were carried out in 3 locations of the homes of the week-long measurement program.
Weekly average as well as instantaneous RSP levels indoors had a wide range and frequently exceeded typical outdoor RSP levels. Elevated indoor RSP levels were clearly associated with tobacco consumption. A considerable variation within homes was observed in the instantaneous RSP levels which were simultaneously measured in different locations of the home. In the regression analysis with weekly average RSP levels in the living room as dependent variable, about 50 % of the variance in RSP levels between homes could be explained by cigarette and cigar consumption and by indicators of occupant activities.The use of ventilation provisions in the living room was positively associated with weekly average RSP levels, which again suggests that occupants tend to increase ventilation when they percieve the pollutant levels as high.
In the homes of the repeated measurement program, the indoor weekly average RSP levels in the 2 smoker's homes were consistently higher (on average by about 60 $\mu g/m^3$), than the levels in the 2 non-smoker's homes. In the non-smoker's homes, highest RSP levels were observed during the stay of smoking

guests. The results of the repeated RSP measurements indicated that the indoor RSP levels were rather stable. This observation is in good agreement with results of another Dutch study in which RSP levels were determined in homes.
Values of the US primary standard for outdoor particulate matter, respectively the proposed revision of the standard, were regularly exceeded in the homes of the week-long measurement program.

Chapter 6 presents the results of the VOC measurements in the week-long and repeated measurement program. In contrast to the other pollutants under study, week-long VOC levels were obtained in 3 age-groups of homes:pre-war homes, post-war homes and less than 6 years old homes. Repeated VOC measurements were carried out in 4 homes and, in addition, in 11 newly built homes before and during occupation.
Most of the 45 VOC, which could be determined quantitatively, were detected in a vast majority of the homes in a wide concentration range. Virtually all VOC concentrations were higher indoor than outdoors. The indoor levels of several of the solvent type of compounds were highly associated with each other in the homes of the week-long measurement program. This suggests the existence of some common sources for these compounds. The nature of these sources (building materials or consumer products), however, is largely unknown yet. The use of solvent and paint during the measurement period and the presence of smokers were significantly associated with the levels of straight-chain and aromatic hydrocarbons, but explained only a minor fraction of the variance in these pollutant levels.
In the 4 homes of the repeated measurement program, fairly stable indoor levels were only observed for some compounds in some homes. In the 11 newly built homes, straight-chain and aromatic hydrocarbon levels were generally higher before and during the first 3 months of occupation than the levels measured in the homes of the week-long measurement program. The application of floor- and wall covering materials gave transient concentration peaks for the non-chlorinated compounds. It was concluded that most VOC have both consumer products and building materials as indoor sources. The highest observed VOC levels in occupied homes were probably the result of occupant activities. At present, there is little information available to evaluate the health risks associated with the observed indoor VOC levels.

Chapter 7, gives a general discussion on the study. It contains some reflections on the study design and interpretation of the results. The appropriateness of comparing indoor pollutant levels with air quality standards for outdoor pollutants is discussed and some implications of indoor air pollution for environmental epidemiology are considered. In the final section of chapter 7, indoor air pollution and public policy are discussed.

SAMENVATTING.

Dit proefschrift behandelt luchtverontreiniging in Nederlandse woningen van uit het oogpunt van volksgezondheid. Het bestaat uit 2 delen. Deel 1 beschrijft de geschiedenis en de achtergronden van het onderzoek naar binnenluchtverontreiniging, en tevens de opzet van het onderzoek in Nederlandse woningen. In deel 2 worden de resultaten van het onderzoek gepresenteerd en besproken.

Deel 1

In hoofdstuk 1 wordt aangegeven om welke redenen de kwaliteit van de binnenlucht aandacht verdient: de campagnes ter beperking van het huishoudelijk energie verbruik, gestart na de "olie crisis" van 1973, de introductie van nieuwe materialen en chemicaliën in de woning en tenslotte de tijdsbestedingspatronen van de mens in westerse geïndustrialiseerde samenlevingen. Dit hoofdstuk introduceert het begrip integrale persoonlijke blootstelling, dat het belang van de woning voor blootstelling aan luchtverontreiniging illustreert. De achtergronden van het onderzoek worden besproken en het onderzoeksdoel wordt geformuleerd: de karakterisering van kenmerkende luchtverontreinigingsniveaus in Nederlandse woningen, in relatie tot eigenschappen van de woning en haar bewoners, vanuit het oogpunt van integrale persoonlijke blootstelling en volksgezondheid. Het laatste deel van hoofdstuk 1 geeft een beknopte beschrijving van de verontreinigingen waarop het onderzoek betrekking had: koolmonoxide (CO), stikstofdioxide (NO_2), inadembaar stof (RSP) en vluchtige organische verbindingen (VOC).
Hoofdstuk 2 geeft een overzicht van de onderzoeksmethoden voor binnenluchtverontreiniging. Typerend voor het onderzoek op dit gebied blijkt te zijn dat het voort komt uit verschillende vakgebieden. In de onderzoeksbenaderingen zijn drie hoofdstromingen onderscheiden en beschreven: de mechanistische, de 'case study' en de 'survey' benadering.
De stand van wetenschap op het gebied van binnenluchtverontreiniging in het algemeen en meer in het bijzonder voor de situatie in Nederland wordt beschreven. Geconcludeerd wordt dat de huidige kennis,met name ten aanzien van de Nederlandse situatie, beperkt is. De huidige inzichten rechtvaardigen bezorgdheid over de kwaliteit van de lucht in Nederlandse woningen.
Hoofdstuk 3 beschrijft de onderzoeksopzet, die bestaat uit 3 meetprogramma's. Deze zijn samengevat in de tabel op de volgende pagina:

Samenvatting van de onderzoeksopzet:

gemeten stoffen	'real-time' metingen	meetprogramma's weekgemiddelde metingen	herhaalde metingen
CO	Wageningen - 12 huizen - winter 82/83, 83/84 - k, w, s*	Ede - 174 na-oorlogse huizen - winter 81/82 - k, w* Rotterdam (binnenstad) - 102 voor-oorlogse huizen - winter 82/83 - k, w*	
NO_2	Wageningen - 12 huizen - winter 82/83, 83/84 - k, w, s*	Ede - 174 na-oorlogse huizen - winter 81/82 - k, w, s* Rotterdam (binnenstad) - 102 voor-oorlogse huizen - winter 82/83 - k, w, s*	Wageningen e.o. - 15 huizen - maart 82-febr.83 - k, w, s*
RSP		Ede - 174 na-oorlogse huizen - winter 81/82 - w* Rotterdam (binnenstad) - 102 voor-oorlogse huizen - winter 82/83 - w*	Wageningen e.o. - 4 huizen: 2 niet-rokers 2 rokers - juli 82-febr.83 - w*
VOC		Ede - 174 na-oorlogse huizen - winter 81/82 - w* Rotterdam - 102 voor-oorlogse huizen - winter 82/83 - w* Ede - 103 huizen ⩽ 6 jaar oud - winter 82/83 - w*	Wageningen e.o. - 4 huizen: 2 niet-rokers 2 rokers - juli 82-febr.83 - w* Ede - 11 nieuwbouw huizen - zomer en herfst 83 - w*

* k = keuken; w = woonkamer; s = slaapkamer

De instrumentatie, de selectie van de in het onderzoek betrokken woningen, de uitvoering van de metingen en de wijze van gegevensverwerking van de drie meetprogramma's worden in hoofdstuk 3 beschreven.

Het doel van het programma van 'real-time' metingen was het bepalen van het concentratieverloop van CO en NO_2 in de woning, het bepalen van de verspreiding van de verontreinigingen door de woning en het bepalen van de dagelijkse variatie in de verontreinigingsniveaus. Ter bepaling van de keukenventilatie in dit meetprogramma werden tracer gas experimenten uitgevoerd met zwavelhexafluoride (SF_6); het gebruik van afvoerloze gasapparaten werd bepaald met behulp van thermokoppels.
Het doel van het programma van weekgemiddelde metingen was het bepalen van de concentratieniveaus van CO, NO_2, RSP en VOC in een groot aantal verschillende woningen. Naast concentratiemetingen werden in dit programma tracer gas bepalingen uitgevoerd in keukens en werd aanvullende informatie over de woning en over de activiteiten van de bewoners verzameld m.b.v. inspectielijsten, enquêtes en dagboekformulieren.
Het doel van het programma van herhaalde metingen was het bepalen van de variatie in weekgemiddelde concentratieniveaus in verschillende seizoenen.

Deel 2

Hoofdstuk 1 beschrijft de indeling van het tweede deel van het proefschrift.
Hoofdstuk 2 beschrijft algemene kenmerken van de woningen die onderzocht werden. Het gebruik van bronnen van binnenluchtverontreiniging, het gebruik van ventilatievoorzieningen en binnendeuren, en de resultaten van de tracer gas experimenten worden (voor zover van toepassing) behandeld. De nadruk ligt op de presentatie van die gegevens over de woning en bewonersactiviteit die in de volgende hoofdstukken gebruikt worden in de gegevensverwerking om de variantie in de gemeten verontreinigingsniveaus te verklaren. Voor de 12 huizen van het programma van de 'real-time' metingen worden voorbeelden gepresenteerd van gegevens-plots van het gebruik van gasapparaten.
In deze huizen werd het kooktoestel 2 tot 6 keer per dag gebruikt; de geiser, voor zover aanwezig, werd 10 tot 35 keer per dag gebruikt. Er was relatief weinig variatie in het gebruik van gasapparaten op verschillende dagen in hetzelfde huis, maar er waren grote verschillen in het gebruik van gasapparaten in verschillende huizen. De resultaten van de tracer gas experimenten wezen op niet-homogene menging van de lucht in de keuken. De 4-uurs gemiddelde waarden van het ventilatievoud en de tracer gas transfer index varieerden sterk zowel binnen een huis als tussen verschillende

huizen.
De positieve respons in het programma van weekgemiddelde metingen was redelijk goed; meer dan 50% van de bewoners waarmee contact gelegd kon worden was bereid om aan het onderzoek mee te werken. In de binnenstad van Rotterdam was echter het percentage bewoners waarmee contact gemaakt kon worden, laag. Huishoudens waarvan alle leden een volledige baan hadden waren vermoedelijk ondervertegenwoordigd in het onderzoek. Vergelijking van de verdeling van woningkenmerken, zoals eigendomsverhouding, woningtype, kookbrandstof, warmwatervoorziening en ruimte verwarming, in de huizen van het programma van weekgemiddelde metingen met gegevens uit verschillende bronnen, bracht geen duidelijke afwijkingen aan het licht ten opzichte van dit referentie materiaal.
Wel werden er grote verschillen aangetroffen in de verdeling van woningkenmerken over de drie leeftijdsgroepen van woningen die onderzocht werden.
Een grote spreiding werd waargenomen in het gebruik van gasapparaten en ventilatievoorzieningen, en in het tabaksgebruik; ook in de resultaten van de tracer gas experimenten werd een grote spreiding waargenomen. Een belangrijk deel van de respondenten bleek gedurende het stookseizoen geen gebruik te maken van de ventilatievoorzieningen in de woonkamer.

Hoofdstuk 3 behandelt de resultaten van de CO-metingen in de programma's van 'real-time' en weekgemiddelde metingen en presenteert tevens resultaten van twee verwante studies.
Eerst worden voor de 12 huizen van het programma van 'real-time' metingen enige kwalitatieve observaties beschreven, die gebaseerd zijn op gegevensplots.
De gemiddelde CO-concentratie over de gehele meetperiode in deze huizen was laag: 0-3 mg/m^3 hoger dan in de buitenlucht. Tijdens het gebruik van afvoerloze gasapparaten werden piekconcentraties van enkele tientallen mg/m^3 CO waargenomen.
In het algemeen was het concentratieverloop van CO in de woon- en slaapkamer vlakker dan in de keuken. Uit het concentratieverloop van CO kon opgemaakt worden dat het roken van tabak de CO niveaus verhoogde met een paar mg/m^3. De marge tussen de door de Gezondheidsraad voorgestelde grenswaarden van CO in de buitenlucht en de waargenomen CO-niveaus in de woningen van het programma van 'real-time' metingen was, voor zover aanwezig, klein.
In de huizen van het programma van weekgemiddelde metingen werden CO niveaus gemeten die dezelfde spreidingsbreedte hadden als de gemiddelde CO niveaus in de huizen van het programma van 'real-time' metingen. Meervoudige regressie analyse met de resultaten van het programma van weekgemiddelde metingen liet een samenhang zien van verhoogde CO niveaus in de keuken met de aanwezigheid van afvoerloze geisers, het brandertype van de

geiser, het gebruik van het gaskooktoestel en met het roken van tabak. Er was een positieve samenhang tussen het CO niveau in woonkamer met het CO niveau in de keuken en met het roken van tabak. Het CO niveau in de woonkamer hing negatief samen met het aantal deuren tussen woonkamer en keuken. Met de regressie analyse kon slechts een gering percentage van de variantie in de CO niveaus in keuken en woonkamer verklaard worden. Het waargenomen effect van de geiser en van het brandertype van de geiser op de CO niveaus in de huizen van het programma van weekgemiddelde metingen kwam goed overeen met de resultaten van een verwante studie naar de CO productie van geisers onder gecontroleerde omstandigheden.
De resultaten van een verwante studie naar COHb niveaus wezen uit dat, onder normale omstandigheden, belangrijke verhogingen (uit medisch oogpunt) van het COHb niveau door het gebruik van afvoerloze geisers met een hoge CO productie, minder vaak voorkomen dan dergelijke geisers zelf voorkomen in het Nederlandse woningbestand.

Hoofdstuk 4 behandelt de resultaten van de NO_2-metingen in de programma's van 'real-time', weekgemiddelde en herhaalde metingen; ook hier worden resultaten van enkele verwante studies weergegeven.
Na het beschrijven van enkele kwalitatieve observaties, gebaseerd op gegevens-plots van de huizen van het programma van 'real-time' metingen, worden de in deze huizen gemeten 1-minuuts, 1-uurs en 24-uurs gemiddelde NO_2 concentraties en het gemiddelde NO_2 niveau over de hele meetperiode behandeld. In het algemeen waren de NO_2-niveaus hoger in de binnenlucht dan in de buitenlucht. In verschillende huizen werden in alle drie de vertrekken 1-uurs gemiddelde piek concentraties gemeten van enkele honderden $\mu g/m^3$ NO_2.
De maximum 1-uur gemiddelde NO_2 concentratie in een huis was in de keuken gemiddeld 6 keer zo hoog als het gemiddelde NO_2-niveau in de keuken over de hele meetperiode; in woon- en slaapkamer was het maximum 1-uurs gemiddelde NO_2-niveau 6 respectievelijk 4 maal zo hoog als het gemiddelde niveau over de hele meetperiode. Het dagelijks maximum van 1-uurs gemiddelde NO_2-niveaus en het 24-uurs gemiddelde NO_2-niveau varieerde aanzienlijk van dag tot dag. Op sommige dagen werden in enkele huizen in woon- en slaapkamer NO_2 piekniveaus gemeten die net zo hoog waren als de piekniveaus die gelijktijdig in de keuken voorkwamen; op andere dagen werden in dezelfde huizen de NO_2-niveaus in woon- en slaapkamer nauwelijks door de NO_2-niveaus in de keuken beïnvloed .
Ook in de huizen van het programma voor weekgemiddelde metingen waren de NO_2-niveaus vaak hoger dan in de buitenlucht. De gemeten NO_2-niveaus kwamen goed overeen met de NO_2-niveaus zoals die gemeten zijn in 4 verwante studies in 700 andere Nederlandse woningen. In de binnenstads woningen van Rotterdam waren de NO_2-niveaus in het algemeen hoger dan in woningen in

kleine stedelijke en plattelands gemeenten.
Ongeveer 50-70% van de variantie in de NO_2-niveaus in de huizen van het programma van weekgemiddelde metingen kon verklaard worden met behulp van meervoudige regressie-analyse. De aanwezigheid van afvoerloze geisers, aanwezigheid en gebruik van gas-kooktoestellen, het NO_2-niveau in de buitenlucht, het type ruimteverwarming en het gebruik van ventilatievoorzieningen hadden een positieve samenhang met de NO_2-niveaus in de keuken. Hoewel er geen directe samenhang aangetoond kon worden, wezen de resultaten van de gegevensverwerking erop dat ook de gebruiksduur van de geiser positief samenhing met het NO_2-niveau in de keuken. Een gunstig effect van het gebruik van een afzuigkap op het NO_2-niveau in de keuken werd alleen gevonden met de gegevens van de woningen in Ede. In 2 van de verwante studies werd een soortgelijke samenhang aangetroffen tussen de aanwezigheid van een afzuigkap en de NO_2-niveaus in de keuken. Het is onduidelijk of de samenhang tussen de variabele 'soort ruimte verwarming' en de NO_2-niveaus betekent dat gashaarden een bron van NO_2 zijn, of dat deze variabele het effect van een ander kenmerk van de woning weergeeft.
De positieve samenhang tussen het gebruik van ventilatievoorzieningen en de NO_2-niveaus in de keuken zou kunnen betekenen dat bewoners extra gaan ventileren als zij de verontreinigingsniveaus als hoog ervaren.
In woonkamers van de huizen in het programma van weekgemiddelde metingen kan 60% van de variantie in NO_2-niveaus verklaard worden met de NO_2-niveaus in de keuken, het aantal deuren tussen keuken en woonkamer, en het type ruimteverwarming. Indien de variabele 'tracer gas transfer index' aan het regressienmodel werd toegevoegd kon 73% van de variantie in NO_2-niveaus in de woonkamer worden verklaard.
Ongeveer 50% van de variantie van de NO_2-niveaus in de slaapkamer kon verklaard worden met behulp van de NO_2-niveaus in andere vertrekken en in de buitenlucht en het aantal deuren tussen keuken en slaapkamer. Na standaardisatie voor deze variabelen waren de NO_2-niveaus in de slaapkamers van Rotterdamse huizen significant hoger dan die in Edese woningen.
In de 3 vertrekken van de huizen van het programma van herhaalde metingen was de variatie in weekgemiddelde NO_2-niveaus gering in de winter, het voorjaar en de herfst, maar groter in de zomer. In de keukens van deze huizen waren de NO_2-niveaus het hoogst gedurende de winter en het voorjaar en het laagst gedurende de zomer. In de andere vertrekken werden geen duidelijke seizoensinvloeden waargenomen. De resultaten van het programma van herhaalde metingen wezen uit dat, gedurende het grootste deel van het jaar, de NO_2-niveaus in verschillende woningen consequent van elkaar verschilden; gedurende het stookseizoen kunnen dergelijke verschillen tussen huizen met (een) enkele meting(en) bepaald worden.
De resultaten van de programma's van 'real-time' en weekgemiddelde metingen toonden aan dat de voorgestelde grenswaarden voor NO_2 in de buitenlucht

veelvuldig overschreden worden door de NO_2-niveaus in keukens; ook in woon- en slaapkamer worden de grenswaarden regelmatig overschreden.

Hoofdstuk 5 behandelt de resultaten van de RSP-metingen in de programma's van weekgemiddelde en herhaalde metingen. Naast de metingen van weekgemiddelde RSP-niveaus werden in 3 vertrekken van de huizen van het programma van weekgemiddelde metingen ook momentane RSP metingen uitgevoerd.
De weekgemiddelde zowel als de momentane RSP-niveaus hadden een grote spreiding binnenshuis en waren vaak hoger dan de niveaus zoals die in de buitenlucht voorkomen. Verhoogde RSP-niveaus hingen duidelijk samen met het roken van tabak. De momentane RSP-niveaus die gelijktijdig in verschillende vertrekken van één woning werden gemeten, varieerden vaak aanzienlijk. Ongeveer 50% van de variantie in weekgemiddelde RSP-niveaus in de woonkamer kan verklaard worden in de regressie-analyse door het aantal gerookte sigaretten en sigaren en door indicatoren die bewoners activiteiten weergeven. Het gebruik van ventilatievoorzieningen hing positief samen met de weekgemiddelde RSP-niveaus, wat er opnieuw op wijst dat bewoners de ventilatie verhogen als zij de verontreinigingsniveaus als hoog ervaren.
In de huizen van het programma van herhaalde metingen waren de weekgemiddelde RSP-niveaus in de 2 huizen met rokende bewoners consequent hoger (ongeveer 60 $\mu g/m^3$ hoger), dan in de 2 huizen met niet-rokende bewoners. In de huizen van niet-rokers werden de hoogste RSP-niveaus gemeten gedurende het verblijf van rokende gasten. De resultaten van de herhaalde RSP-metingen wezen uit dat binnenshuis de RSP-niveaus tamelijk constant waren. Dit komt goed overeen met resultaten van een andere Nederlandse studie waarin RSP-niveaus binnenshuis gemeten zijn. De waarden van de Amerikaanse norm voor zwevend stof in de buitenlucht, respectievelijk van de voorgestelde wijziging van die norm, werden regelmatig overschreden in de huizen van het programma voor herhaalde metingen.

Hoofdstuk 6 behandelt de resultaten van de VOC-metingen in de programma's van weekgemiddelde en herhaalde metingen. In tegenstelling tot de andere verontreinigingen zijn de weekgemiddelde VOC-niveaus bepaald in 3 leeftijdsgroepen van woningen: in vooroorlogse, in na-oorlogse en in na-oorlogse woningen van minder dan 6 jaar oud. Herhaalde metingen werden uitgevoerd in 4 huizen, en bovendien in 11 nieuwbouw woningen, vóór en tijdens bewoning. De meeste van de 45 VOC's die kwantitatief bepaald konden worden, werden in een grote meerderheid van de woningen aangetroffen, in concentraties die een grote spreiding vertoonden. Vrijwel alle VOC-niveaus waren binnenshuis hoger dan buitenshuis. De niveaus van verschillende oplosmiddelachtige verbindingen vertoonden een sterke samenhang in de woningen van het programma van weekgemiddelde woningen. Dit wijst er op dat er voor deze verbindigen enkele gemeenschappelijke bronnen zijn. De aard van deze

bronnen (bouwmaterialen of consumentenartikelen) is echter nog grotendeels onbekend. Het gebruik van oplosmiddelen en verf en de aanwezigheid van rokers hing significant samen met de niveaus van onvertakte en aromatische koolwaterstoffen, maar verklaarde slechts een gering percentage van de niveaus van deze verontreinigingen.
In de 4 huizen van het programma van herhaalde metingen werden slechts voor enkele verbindingen in een enkel huis min of meer stabiele VOC-niveaus gemeten. Vóór en gedurende de eerste 3 maanden van bewoning waren de niveaus van onvertakte en aromatische koolwaterstoffen in het algemeen hoger in de 11 nieuwbouw woningen dan in de woningen van het programma van weekgemiddelde metingen. Het aanbrengen van vloer- en muurbedekkingsmaterialen ging gepaard met een tijdelijke verhoging van de niveaus van ongechloreerde verbindingen. Geconcludeerd werd dat de meeste VOC's binnenshuis zowel consumentenartikelen als bouwmaterialen als bron hebben. De hoogste gemeten VOC-niveaus in bewoonde huizen waren waarschijnlijk het gevolg van activiteiten van de bewoners. Tot op heden is er vrijwel geen informatie voorhanden voor de evaluatie van eventuele gezondheidsrisico's van de waargenomen VOC-niveaus binnenshuis.

Hoofdstuk 7 geeft een algemene discussie over het onderzoek. Dit hoofdstuk bevat een aantal overwegingen over de onderzoeksopzet en over de interpretatie van de resultaten. De geschiktheid van grenswaarden voor verontreiniging van de buitenlucht voor de evaluatie van de gemeten binnenluchtverontreinigingsniveaus wordt besproken en enkele consequenties van binnenluchtverontreiniging voor het milieu-epidemiologisch onderzoek worden behandeld. In het laatste gedeelte van hoofdstuk 7 wordt het (overheids) beleid ten aanzien van binnenluchtverontreiniging besproken.

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APPENDIX A. Tracer gas experiments to evaluate ventilation and air infiltration

Introduction

The process of air infiltration (uncontrolled leakage of air through cracks in the building envelope) and ventilation in buildings has been investigated extensively during the past decades, but is still poorly understood. In their literature survey on air infiltration in buildings, Ross and Grimsrud (1978) quote from unpublished material of the Ohio State University, USA (reprinted in an appendix of their report) a list of 15 significant parameters which are required to analyse the infiltration and ventilation phenomena.
The most important parameter affecting the ventilation of homes (and the least understood), is the activity of occupants; few studies have addressed the effect of this parameter on the ventilation of homes. Consequently, models which adequately describe ventilation in occupied homes do not exist up to now.

Direct methods to determine the ventilaion of a room are the tracer gas techniques such as the rate-of-decay method and the constant flow and constant concentration method (Sherman et al. 1981). These techniques involve the emission of a tracer gas in the room and the subsequent measurement of the tracer gas concentration in the room; then, the ventilation of the room can be calculated from a mass balance equation. The ventilation of a room is commonly expressed as the air change rate per hour (ACR): the number of times per hour the air in the room is completely refreshed. An ACR of 2 (h^{-1}) in a room with a volume (V) means that an air volume of 2 times V has passed the room in 1 hour. In general, when the ventilation of a room is expressed as ACR, conditions of ideal mixing of the air in the room are required and the volume of the room must be known.
To evaluate the ventilation of the kitchen and the transfer of kitchen air to the living room in occupied homes in a survey type of study, the constant flow method is most appropriate. With this method, average ventilation rates can be determined over time periods of several days or longer. The measurements can be carried out with relatively simple and cheap equipment, which can be left unattended over periods of a week or longer (Sherman et al. 1981).
With the constant flow method, tracer gas is emitted in a room of volume V (m^3) with a known and constant flow, P (mg/h), during the entire measurement period, and the average tracer gas concentration during the measurement period, C (mg/m^3) is determined in the room.
Under conditions of ideal mixing of the air in the room, the average ACR

can be calculated from formula 1.

$$ACR = \frac{P}{V.\bar{C}} \ (h^{-1}) \qquad (1)$$

A disadvantage of the use of ACR to describe the ventilation of a room in occupied homes is that conditions of ideal mixing are seldom met. The effective volume of air which participates in the air exchange is therefore not always the same as the physical volume of the room. Air in corners and under the ceiling may not communicate well with the rest of the air in the room, and doors to other parts of the home may be closed or open during the measurement period. The effective room volume is therefore difficult to assess. Furthermore, the air exchange between 2 rooms, i.e. between kitchen and living room, cannot be described easily in terms of ACR.

A quantity to describe the ventilation conditions in a room, or between rooms, which does not suffer from the uncertainty about the effective room volume is the transfer index (TI).
The TI describes the exposure to a tracer gas at a certain point, after release of the tracer gas at another point in the room, or in another room. In other words, it expresses the efficiency in protecting a given position from exposure to airborne contaminants, released at another point in the ventilation system (Lidwell 1960).
With the constant flow method, the average TI can be calculated from formula 2.

$$TI = \frac{\bar{C}}{P} \ (h/m^3) \qquad (2)$$

Thus, the higher the ventilation of the room and the larger the room volume, the lower the TI will be. Under conditions of ideal mixing and a known effective room volume, the TI is the reciprocal of the product of the ACR and effective volume V.

A disadvantage of the TI is that it describes the ventilation relationship between 2 points in a ventilation system. Under conditions of incomplete mixing, the TI therefore depends on the choice of the tracer gas emission and sampling point. In addition, the concept of the TI with its dimension of the reciprocal of volume per time unit, is more difficult to interprete than the more current concept of ACR with the dimension of air-changes per hour, in particular for people who are not familiar with typical values for the volume of rooms in Dutch homes. Therefore, the ACR and TI were both used in this study to evaluate ventilation conditions in the homes.

The tracer gas experiments

In this study, tracer gas experiments in kitchens were incorporated in the real-time monitoring program and in the week-long measurement program. Sulfurhexafluoride (SF_6) was selected as a tracer because it has several properties which make it very suitable for this purpose: SF_6 is not a normal constituent of air, it is stable, not toxic, non-flammable, and it can be measured with high sensitivity and a wide dynamic range by electron capture detectors. In addition it can be monitored continuously by an infrared spectrophotometer. Furthermore, equipment and expertise for routine measurements of SF_6 concentrations was available.

In the real-time measurement program, SF_6 was emitted in the kitchen and the SF_6 concentration in the kitchen was monitored continuously by an infrared spectrophotometer. In the week-long measurement program, the tracer gas was emitted in the kitchen and time-integrated samples were taken in kitchen and living room to determine the average SF_6 concentration in the 2 rooms. The emission and sampling point in the kitchen were located at breathing height, at opposite sides of the kitchen at places where the air was considered to communicate well with the area in which the occupants would move. Corners, alkoves and locations near windows, doors, ventilation ducts and -appliances were avoided as well as possible. The sampling point in the living room was located in a similar way. Thus, the TI can be considered as an index of the capacity of the living area in the kitchen and living room, to trap pollutants which were generated in the kitchen.

From the data of the real-time measurement program, kitchen ACR were calculated from the SF_6 emission rate, the 30-minute average SF_6 concentration in the kitchen and the physical volume of the kitchen, using formula 1. In kitchens without an open connection to the living room, the net physical volume was used, by subtracting the volume of large objects like kitchen sink, refrigerator, etc.from the total kitchen volume. In homes with an open kitchen/living room, the physical volume was the sum of kitchen and living room volume.The kitchen ACR were plotted versus time of day for visual inspection of the data. To reduce the amount of data and to smoothen the effect of short-term fluctuations in the SF_6 concentration, 4-hour average ACR and TI were calculated from the 4-hour average SF_6 concentrations in the kitchen, using formula 2.
For the week-long measurement program, average ACR were calculated with formula 1, for the kitchen and for homes with an open kitchen, also for the living room. With formula 2, average TI were calculated for the kitchens and for the living rooms.

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APPENDIX B. NO_2 measurements in homes with the Palmes diffusion tubes

Introduction

Palmes diffusion tubes (Palmes et al. 1976) are used in an increasing number of studies as a convenient sampler for NO_2 indoors. These passive samplers are 8 cm long, 1 cm inner diameter, acrylic tubes with stainless steel wire mesh, coated with the NO_2 absorbent triethanolamine, inserted at the closed end of the tube. Atmospheric NO_2 is transferred from the open end of the tube to the absorbent at the closed end according to Ficks first law (equations), due to molecular diffusion.

$$Q = -D.C.\frac{A}{Z}.t \qquad (1)$$

where Q = quantity of transferred NO_2 (moles)
D = diffusion coefficient of NO_2 in air (cm^2/sec)
C = NO_2 concentration at the open end of the tube (moles/cm^3)
A = cross-sectional area of the tube (cm^2)
Z = the length of the tube (cm)
t = time of exposure of the tube (sec)

After sampling, the absorbed NO_2 is analyzed spectrophotomatically at 540 nm, 30 minutes after adding 2.1 ml Salzman reagent to the tube. NO_2 concentrations can be calculated with equation 1 from the amount of collected NO_2, and the diffusion coefficient (D=0.154 cm^2/sec) of NO_2 in air.

Theoretically, the effect of temperature on the sampling rate is proportional to the square root of T (°K); the mass transfer is insensitive to pressure. Turbulence, due to high air velocities at the open end of the tube may shorten the diffusion path length and increase the sampling rate. According to Palmes et al. (1976), air velocities below 0.5 m/sec have no appreciable effect on the sampling rate.

The diffusion tubes were tested by the U.S. National Bureau of Standards (Cadoff et al. 1979) and by Warren Spring Laboratory in the U.K. (Apling et al. 1979). A lower detection limit of the tubes of about 1200 µg/m^3 over 1 hour sampling period (or 4 µg/m^3 over one week) was established. Accuracy was demonstrated to be better than ±10 %; precision was better than 4 µg/m^3 for a one week sampling period.
Even though the use of the diffusion tubes in indoor air pollution studies is wide spread, only one field-validation study with NO_2 reference measurements has been published to date to our knowledge. In that study, diffusion

tubes were exposed over a one week period in the kitchen of 3 homes. A Bendix NO_x chemiluminescence monitor was used as reference method. The differences in NO_2 concentrations obtained with two methods were within the precision range of the monitor (±7 %) (Appling et al. 1979). Recently Hoen et al. (1984), reported a good agreement between average results of diffusion tubes and continuous monitor. This was, however, not a direct comparison, because the average NO_2 concentrations of the tubes were calculated from a regression model for the periods of heater use.

Since 1979, the NO_2 diffusion tubes were used at the Agricultural University Wageningen in survey type studies on NO_2 pollution in homes. Before and during the use of the tubes in this study laboratory tests with the tubes were carried out and the performance of the tubes was compared with the results of a Bendix chemiluminescence monitor in 10 homes. The laboratory test and the performance test in the homes will be discussed seperately.

Laboratory tests

Experimental

The laboratory tests were carried out in an exposure chamber of 240 liters, in which the test tubes were exposed to NO_2 concentrations of about 300 $\mu g/m^3$ during periods of 20 to 75 hours. The NO_2 concentrations were generated by a dynamic dilution system; pressurized air was passed through columns of activated charcoal and molecular sieve (5A Mesh) and mixed with NO_2 from a NO_2 permeation chamber.
The NO_2 concentration in the exposure chamber was monitored continuously with a Bendix chemiluminescence monitor during the test runs. Relative humidity in the exposure chamber was less than 5 %. In each of the 10 runs a minimum of 10 tubes were exposed.

The tubes were prepared and analysed according to the method described by Palmes. After zero adjustment with zero air from the dilution system, the NO_2 monitor was calibrated against 250 $\mu g/m^3$ NO from a certified cylinder of Spectraseal NO calibration gas and against the known NO_2 concentration in the mixing chamber of the dynamic dilution system. Periodically, the monitor was cross-referenced against other monitors and calibration systems at the laboratory.

In a later phase, the tests were continued with humidity conditioning. In the same experimental design, water was dripped and evaporated in a round-bottom flask, through which the air was passed before it entered the expo-

sure chamber.
Again, the NO_2 concentration in the chamber was monitored with the Bendix NO_x monitor.
A total of 41 succesful humidity conditioned test runs were carried out in the range of < 5-85 % relative humidity, determined in the chamber with a Lambrecht thermohydrograph.

Unfortunately, the humidity conditioned exposure experiments could not be carried out by the same analyst.
The exposure tests without humidity conditoning and the first 12 humidity conditioned runs were carried out by 1 analyst (R.H. range 5-85 %). A second analyst performed 12 succesful humidity conditioned runs in the range of 37-80 % R.H.. The last 17 runs in the R.H. range of 5-50 % were performed by a third analyst.

Results

Tests without humidity conditioning

In the winter of 1981/1982 10 successful tests were carried out. From the known exposure (concentration x time) of the tubes, the experimental diffusion coefficient D was calculated with formula 1; the results are presented in table 1. The coefficient of variation of D over the 10 tubes in each run ranged from 4 to 11 % with an average of 7.5 %. The coefficient of variation of the average D from the different runs was 6 %.
The experimental D with an average value of 0.115 cm^2/sec was consistently lower than the value of 0.154 cm^2/sec used by Palmes as the best estimate for the diffusion coefficient of NO_2 in air.

At the time, no good reasons for the differences between the experimental and theoretical D were found, although such differences in diffusion coefficients derived in different ways are not uncommon (Reid and Sherwood 1966).
During the course of the project, several possible explanations for the different D were brought forward.
First, a temperature effect on the performance of the tubes was reported by Girman et al. (1983). They observed that at 15 °C, the collection efficiency of the tubes was 15 % less than at 27 °C, probably due to a liquid-solid phase transition of the triethanolamine absorbent at 21 °C. During our exposure tests, the temperature in the exposure chamber was always 22 °C or a few degrees higher. A tempertaure effect is therefore not a probable explanation for differences between experimental and theoretical D.

Second, starvation may occur at the open end of the tube at very low air velocities. Under these conditions, an external boundary layer, in which NO_2 transport is controlled by diffusion, will lengthen the diffusion path. With formulas by Tompkins and Goldsmith (1977) and Brown et al. (1981) a starvation error of about minus 12 % was calculated for the experimental conditions during the test runs, with air velocities between 0.05-0.1 cm/sec. This starvation error is not large enough to explain completely the observed differences in experimental and theoretical D.
Third, humidity effect might be responsible for the differences. To our knowledge, tests of the effect of humidity on the performance of the diffusion tubes have not been reported to date. For this reason, the humidity conditioned exposure tests were carried out.
The precision of the tubes did not change under the influence of humidity. The coefficient of variation over the tubes in each run were comparable to those found in the first 15 tests.
To test a possible effect of humidity on the accuracy of the tubes, the data of the three analysts were treated separately. From the results of the first analyst (R.H. range 5-85 %) the following regression line was calculated:

$D = 0.125 + 0.00038$ R.H. (R^2=0.85; F=58.6; d.f.=1, 10; $p < 0.001$)

From the data of the second analyst (R.H. range 37-80 %) no significant association between D and R.H. could be observed. With the data of the third analyst (R.H. range 5-50 %) the calculated regression line was:

$D = 0.116 + 0.00045$ R.H. (R^2=0.60; F =22.1; d.f.=1, 15; $p < 0.001$)

In figure 2 the results of the 3 analysts are plotted together. The regression line fitted through these data was:

$D = 0.120 + 0.00038$ R.H. (R^2=0.65; F =73.4; d.f.=1, 39; $p < 0.001$)

Discussion

The precision of the diffusion tubes in the laboratory tests was comparable with findings reported by Apling et al. (1979) and Cadoff et al. (1979). In dry air, the experimentally derived D of 0.115 cm^2/sec was systematically lower than the value of 0.154 cm^2/sec used by Palmes et al. (1976). Part of this difference may be due to starvation because of low air velocities.
Although the results of the humidity conditioned tests are not considered conclusive because of possible analyst bias, there is the suggestion that a humidity effect on the performance of the tubes does exist.

The important question of course is, how the distribution of differences in air velocity, humidity and temperature in homes will affect the performance of the diffusion tubes for indoor measurements. Changes in temperature from 15-25 °C and in R.H. from 30-80 % between different rooms, and in a single room over a measurement period of several days, are realistic conditions in Dutch homes. Furthermore, little is known about air velocities near the objects and surfaces to which the tubes are normally attached. Stagnant conditions at the sampling location may not be uncommon and starvation may in fact occur in some cases.
It was therefore decided to use the opportunity to compare the performance of the tubes with the results of the real-time monitoring program (cf. part 1, section 3.3 and part 2, section 4.1).

Performance tests in homes

Experimental

In the real-time monitoring program duplicate diffusion tubes were placed near the sampling points in livingroom, bedroom and outdoors. In the kitchen, duplicate tubes were placed near the most central of the 3 sampling points.
It should be noted that the real-time monitoring program was not designed to test the performance of the tubes, but merely offered an opportunity for comparison.

Results

In the field tests, the coefficients of variation of the series of duplicate samples were about the same as in the laboratory tests. The results of the simultaneously measured tube and monitor concentrations are given in table 2. Tube concentrations were calculated with the theoretical diffusion coefficient of 0.154 cm^2/sec.
The overall correlation coefficient for all measurements was 0.93, while the correlation coefficients for the variuos locations varied between 0.83 and 0.95. The correlation was even better when home 8 was considered as an outlier and not taken into account. The overall coefficient of variation between all tube and monitor measurements was 14 %; without home 8, this was 10 %.
In all kitchens, the results of the tubes were higher than those of the monitor. The ratio was on average 1.33. For the living room and bedroom, the ratio averaged 0.88, respectively 0.85. Outdoors, the average was 1.03.

Discussion

One of the explanations for the observed differences in tube/monitor ratios may be an unrecognized systematic bias in the real-time monitoring equipment and sampling arrangements:

- the monitor may underestimate the NO_2 concentration because of quenching by water vapor (Matthews et al. 1977). The high humidity in the kitchen during the use of gas appliances and NO_2 production, might be the reason for the high ratio in the kitchen; other (indoor) locations with lower R.H. would be less affected.
- a higher NO_2 concentration at the most central sampling point in the kitchen where the tubes were placed, than at the other 2 sampling points due to inhomogenicity would also result in a tube/monitor ratio higher than one in the kitchen; the other locations had only one sampling point, adjacent to the tubes.

Another possible explanation for the differences between tubes and monitor is the existence of a temperature, humidity or starvation effect on the tubes. A different distribution of these parameters over the 4 sampling locations would explain the differences in tube/monitor ratios between the locations. It cannot, however, explain the high ratio in the kitchen.

Whatever the reason, or combination of reasons, for the differences between tubes and monitor is, both methods have been commonly used for NO_2 measurements in homes.
The temperature effect reported by Girman et al. (1983), the possible humidity effect and the lack of knowledge about air velocities in homes suggest that the often quoted accuracy figure (< 10 %) from the studies of Apling et al. (1979) and Cadoff et al. (1979) is too optimistic for the use of the tubes in homes.
Further research is needed , in particular on the possible effect of humidity on the performance of the NO_2 diffusion tubes. For the time being, the more conservative theoretical value of the diffusion coefficient of 0.154 cm^2/sec seems the best choice for calculating the results of diffusion tube measurements in field studies.

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Table 1. Laboratory experiments; experimental diffusion coefficients (D) from 10 exposure experiments at a relative humidity of less than 5 %

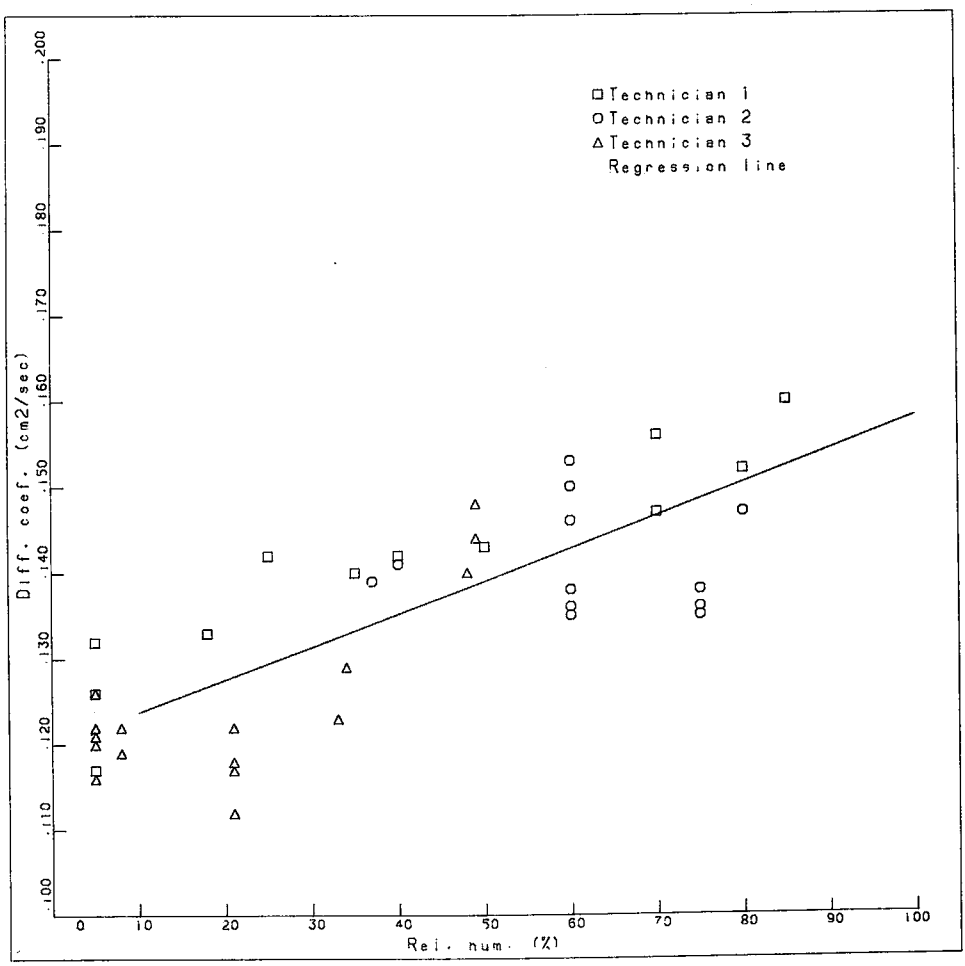
Run	Concentration ($\mu g/m^3$)	Exposure time (h)	D (cm^2/sec)
1	314	45	0.105
2	301	53	0.104
3	310	21.5	0.116
4	308	74	0.106
5	310	73	0.105
6	305	29	0.124
7	306	70	0.115
8	297	48	0.121
9	295	66.5	0.123
10	305	26	0.119

Table 2. NO_2 concentrations (in $\mu g/m^3$) at four sampling locations during 10 periods in 9 homes, measured simultaneously with dupulicate diffusion tubes and a chemiluminescence monitor

home nr. and sampling period		kitchen	living room	bedroom	outdoors
home 2	monitor	188	74	54	42
4 days	tubes	229	56	46	43
	ratio	1.22	0.76	0.85	1.02
home 3	monitor	146	81	53	58
7 days	tubes	185	73	46	68
	ratio	1.27	0.90	0.87	1.17
home 4	monitor	54	43	30	37
7 days	tubes	64	43	25	37
	ratio	1.18	1.00	0.83	1.00
home 5*	monitor	36	32	22	26
12 days	tubes	42	37	25	38
	ratio	1.17	1.16	1.14	1.46
home 6	monitor	52	39	15	50
8 days	tubes	61	40	15	41
	ratio	1.17	1.03	1.00	0.82
home 7*	monitor	93	92	40	76
7 days	tubes	105	90	34	68
	ratio	1.12	0.98	0.85	0.89
home 8	monitor	63	35	34	40
3 days	tubes	116	16	29	40
	ratio	1.83	0.46	0.85	1.00
home 8	monitor	89	39	40	36
5 days	tubes	174	17	25	28
	ratio	1.96	0.44	0.63	0.78
home 9*	monitor	40	33	22	25
4 days	tubes	45	30	22	27
	ratio	1.13	0.91	1.00	1.08
home 10*	monitor	156	116	91	52
8 days	tubes	207	132	44	54
	ratio	1.33	1.14	0.48	1.04

* homes, with open kitchen/living room

FIGURE 1. Experimental diffusion coefficients calculated from the results of 41 humidity controlled laboratory experiments at various relative humidities.



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Curriculum Vitae

Erik Lebret was born on April 2nd, 1953 in Middelburg, where he attended primary and secondary school. He studied Environmental Sciences at the Wageningen Agricultural University from 1971 until 1980, with majors in Air Pollution, Environmental Health and Management Studies. After his study, he joined the Department of Air Pollution and the Department of Environmental and Tropical Health of the Wageningen Agricultural University, where he carried out the study which is described in this thesis. In 1985, he received a grant from the Netherlands organization for the advancement of pure research (ZWO), for a 10 month stay at the Harvard School of Public Health, Boston, USA.