1985

INDOOR AIR POLLUTION IN THE NETHERLANDS

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INTRODUCTION

A major part of exposure of people to common air pollutants such as carbon monoxide, nitrogen dioxide and particulate matter is often occurring inside homes. Not only do people in Western industrialized society spend much more time inside their homes (about 70%) than outside, but the concentration levels to which they are exposed indoors are also often higher than outdoors. One problem in assessing dose-response relations for health effects of air pollution lies therefore in the determination of realistic population exposures to a pollutant ; central outdoor monitoring systems are usually insufficient for that purpose.

Already in the sixties Biersteker et al. (1967) suggested that indoor air pollution might be a neglected variable in epidemiology. Since then, studies have indicated that there are probably health effects of indoor pollution by NO_2, formaldehyde and by passive smoking but there are still a lot of uncertainties about the implications of air pollution inside premises on health (NAS, 1981).

At the Wageningen Agricultural University the Department of Air Pollution and the Department of Environmental and Tropical Health started a study in 1980 to characterize air pollution in Dutch homes, as a step toward total exposure assessment. The pollutants of concern are primarily carbon monoxide, nitrogen dioxide and respirable suspended particulate matter (RSP). In cooperation with the National Institute of Public Health, volatile hydrocarbons are also studied.

In this paper we will report some of the results of our studies. The different pollutants will be discussed separately. The data presented here were mainly gathered in 2 surveys: in October - December 1980 the CO production of waterheaters, and NO_2 levels were measured in about 300 houses in Arnhem and Enschede. The second survey was conducted in 175 houses in Ede between October 1981 and April 1982. In Ede weekly average levels of RSP, NO_2 and volatile hydrocarbon were measured; during the days of the measurement occupants kept diaries about the use of ventilation provisions, gas appliances, vacuum cleaning and tobacco smoking. Materials and methods of the

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Arnhem/Enschede survey, and house selection and response figures of both surveys have been described elsewhere (Brunekreef et al, 1982A, Boleij et al, 1982).

CARBON MONOXIDE

Carbon monoxide was one of the first recognised indoor air pollutants in the Netherlands with coal fired space heaters and town gas as important sources. Between 1961 - 1970 over nine hundred fatal CO poisonings were registered. The change to natural gas as domestic energy source in the mid-sixties, improved the situation. However, the geiser, an instantaneous gas-fired waterheater, came up as a new indoor source of CO. This was caused by the type of burner (primary aerated) used in most natural gas geisers; we will refer to this later. This type of waterheater is now present in 70% of the Dutch homes and is usually unvented.

In the Arnhem/Enschede survey we measured the source strength of 254 of these appliances. The results of the CO concentration measurements in flue gases of the geisers are summarized in Table 1 (CO flue gas concentration in ppm, and in mg CO/MJ, assuming a gasinput of 20 l/minute and a CO_2-concentration in flue gas of 6%).

TABLE 1. Frequency distribution of CO-concentration in flue gases (ppm) of geisers and CO source strength (mg/MJ). Indoor air pollution survey Arnhem/Enschede 1980.

CO concentration (ppm)	CO source strength (mg/MJ)	n	%
≤ 100	< 6	119	53
101 - 300	6 - 18	46	20
301 - 600	19 - 36	20	9
> 600	>36	41	18
	missing values	28	
		254	100

In 27% of the cases the level of 300 ppm CO in flue gases was exceeded. This is the level above which the local gascompanies require maintenance.

The source strength of the geisers was largely determined by burnertype and maintenance system. The 300 ppm CO level was exceeded by 32% of the appliances with primary aerated burners. This was only the case for 3% of the secondary aerated burners. The maintenance system proved to be important in reducing the CO source strength of the primary aerated burners (Brunekreef et al, 1982A).

In addition CO levels at breathing height were measured after operating the geiser for 15 minutes. Doors and windows of the kitchens were closed during measurement but permanent ventilation provisions were left open.

In 17% of the cases CO levels above 50 ppm (the installation code standard) were reached in the kitchen.

TABLE 2. Distribution of cases according to CO level at breathing height, burner type, city, presence of a geiser flue. Indoor air pollution survey Arnhem/Enschede 1980.

	bunsen burner				diffusion burner			
	flue		no flue		flue		no flue	
	Arnhem	Enschede	Arnhem	Enschede	Arnhem	Enschede	Arnhem	Enschede
CO ≤ 25 ppm	37	40	45	14	9	4	22	1
CO > 25 ppm	3	0	49	10	0	0	2	1

missing values 17

Linear logit analysis showed a significant effect of the presence of a geiser flue and of burner type on the CO levels in the kitchen. The second-order interaction between city, burner type and CO level is probably due to the better maintenance system in Enschede (Cf. Table 2). Since the measurement conditions were experimental, these were only potential CO levels.

To evaluate CO exposure under normal conditions we proceeded with a pilot study of CO in exhaled breath (Van der Velde and Verhoeff, 1982). Twelve houses were selected on criteria for potentially high CO levels, i.e. presence of an unvented geiser with a primary aerated burner and a CO level in flue gases of 600 ppm or more.
CO concentration in exhaled breath to estimate COHb was measured on two different days among 29 occupants before and after the cooking/dishwashing period, a period of frequent use of the geiser. The participants were asked to exhale through a 250 ml glass tube after holding their breath for 20 seconds. The tubes were sealed after complete exhalation, and subsequently analyzed by GLC. COHb values were calculated using the equations by Peterson (1970) for non-smokers and Jarvis (1980) for smokers.

With one exception, the COHb levels of 12 non-smokers were all below 3%, the level on which the Dutch ambient air quality standard for CO is based. Only small and insignificant COHb increases were found after the cooking/dishwashing period (Table 3).

TABLE 3. % COHb before and after cooking and dishwashing.
Number of participants in parentheses.

	day 1		day 2	
	non-smokers	smokers	non-smokers	smokers
before	0.24 (12)	3.76 (17)	0.17 (10)	3.22 (15)
after	1.29	4.34	0.69	3.59
two sample t-test	>.05	>.05	>.05	>.05

The findings of this study suggest, that under normal conditions of use, the probability of high CO uptake in these kitchens is small. Nevertheless there are still several fatal geiser related CO poisonings each year.

NITROGEN DIOXIDE

NO_2 is generated indoors by combustion processes in unvented appliances. In the Netherlands these are the cooking range, oven and geiser. Results from NO_2 emission studies of gas appliances in the United States (Dewerth, 1974, Cote et al, 1974, Traynor et al, 1979) range from 4.5 - 10 cm³ NO_2/MJ for range burners and 3 - 7 cm³ NO_2/MJ for ovens. From figures reported by Takens (1972) and Van de Linden et al, (1969) NO_2 emission factors can be calculated of 3 cm³/MJ for ovens and 2 cm³/MJ for geisers. Based on our own measurements of NO_2 production by geisers, we arrive at a source strength of 2.5 cm³ NO_2/MJ.

To measure the actual NO_2 levels that exist in Dutch houses we used Palmes diffusion tubes in the surveys of Arnhem/Enschede and Ede. Weekly average NO_2 concentrations were measured in kitchen and living room and in Ede also in one bedroom per house. The frequency distribution of the results from the survey in Ede is given in Figure 1. The corresponding weekly mean outdoor concentrations averaged 44 µg/m³ (range 25 - 72 µg/m³).

A preliminary stepwise multiple regression analysis of the NO_2 levels in the kitchen in Ede with simple house characteristics, resulted in a model containing five independent variables (Table 4).

ABLE 4. Regression coefficients and Beta values of regression model with the logarithm of the NO_2 levels (ppb) in the kitchen as dependent variable. Indoor air pollution survey Ede, winter 81/82.

ndependent variable	Regression coefficient	Beta
utdoor NO_2 level (ppb)	.007	.15
resence of unvented geiser (no=1, yes=2)	.31	.49
ooking fuel (electricity=1, gas=2)	.29	.20
itchen volume (m³)	-.001	-.19
resence central heating system (no=1, yes=2)	-.13	-.18
onstant	.8	
5,155 = 46.2 ; $p < .0001$;		

he model explained 60% of the variance in the logarithm of the NO_2 concentration in he kitchen. The impact of geiser and cooking fuel on the NO_2 concentration is emonstrated in Table 5, assuming a kitchen volume of 15 m³, an outdoor NO_2 level of 4 μg/m³ and assuming absence of central heating system (values predicted by the model).

ABLE 5. Predicted kitchen NO_2 levels (μg/m³)

	Electric cooking	Gas cooking
No unvented geiser	49	94
Unvented geiser	100	194

In the living room, 68% of the variation in the log NO_2 level among houses was ccounted for by the log NO_2 level in the kitchen, the number of doors between kitchen nd the room and by the type of space heating. Simularly 59% of the variation in bed-oom concentrations could be explained by these variables, in combination with the utdoor and living room concentration.

The NO_2 concentration levels we found in Ede, as well as the factors influencing hese levels are in line with the earlier findings in the Arnhem/Enschede survey Boleij et al, 1982).

contrast to CO, there are no regulations for NO_2 as an indoor pollutant. ccording to the gas installation code, however, CO_2 concentration of 1% is allowed n kitchens. Given an emission ratio of 10^{-4} mg NO_2/mg CO_2 for geisers, levels of about .8 mg/m³ (1 ppm) NO_2 may occur at this level. If we compare the observed weekly verage NO_2 levels with the proposed Dutch NO_2 standard for ambient air, assuming a ne hour peak to weekly mean ratio of 5, it is clear thet the 99.7 percentile of 50 μg/m³ (24 h average), as well as the 99.99 percentile of 300 μg/m³ (1 h average) s frequently exceeded in Dutch houses.

ESPIRABLE SUSPENDED PARTICULATE MATTER

Respirable suspended particulate matter (RSP) is a type of air pollution with a ide range of sources. Several studies have indicated higher indoor than outdoor oncentration of RSP, clearly relating high indoor concentrations with the presence f smokers in the house. In the survey in Ede we measured weekly mean RSP concentration n living rooms.

Samples were collected on glasfibre filters with a flow of 2 l/min. The non-respira-le fraction was removed by Casella cyclones (2.5 μm 50% cut-off). The sampling units ere placed in the living room at approximately breathing height. Duplicate samples aken in 8 houses, indicated a coefficient of variation of 5% for sampling plus analy-is. The frequency distribution of the measured RSP concentration is given in Figure 2.

The geometric mean concentration was 61 $\mu g/m^3$.

To explain differences in RSP concentration between houses, several house and household characteristics were used in a multiple regression analysis. In some cases a logarithmic transformation of a variable was carried out after inspection of the variable distribution and of the residuals of the models. The variables selected in the regression model were the logarithm of the number of cigarettes smoked and the total of person-hours spent in the house during the measurement period as an indicator for indoor activity. Together these variables accounted for 55% of the variation in the logarithm of the RSP concentration:

log RSP conc. = 1.29 + 0.3 (log N cig.) + 0.025 (100 person hours)

F 2,151 = 90.5 ; $p < .0001$). With a mean of 420 person-hours, a background concentration can be calculated from the model of 25 $\mu g/m^3$ if no cigarettes were smoked. Other variables such as ventilation system, cooking or heating fuel, frequency of vacuum cleaning and the use of ventilation provisions, could not significantly improve the model.

The influence of smoking on the RSP concentration is consistent with the findings of others. Dockery and Spengler (1981) reported a concentration increase of 0.9 $\mu g/m^3$ per cigarette in not fully air conditioned houses. From our model we calculate a mean impact of 0.6 $\mu g/m^3$ per cigarette when 20 cigarettes per day are smoked and of 1.8 $\mu g/m^3$ per cigarette when 20 cigarettes per week are smoked. Given differences in house design, ventilation habits, etc., these findings are fairly close. Moschandreas (1978) also found higher concentrations in houses with children of 7 years or younger, which might reflect the additional influence of indoor activity on RSP concentration levels.

The concentrations measured in Ede only covered one week per house in the winter period. Therefore we cannot compare the results with the annual mean value - 75 $\mu g/m^3$ - of the US Ambient Air Quality Standard for total suspended particulate matter.

Preliminary results from a program in which we measure RSP concentration every second week in 2 smokers and 2 non-smokers homes and the long-term average concentrations of suspended particulate matter measured by Brunekreef and Boleij (1982) in Dutch houses, suggest, however, that the annual mean value of 75 $\mu g/m^3$ will be exceeded regularly in Dutch houses.

VOLATILE HYDROCARBONS

The interest in volatile hydrocarbons indoors was triggered in the Netherlands by cases of soil pollution. Volatile hydrocarbons from chemical spills in the soil were found to penetrate into houses. Reference material was badly needed to interprete findings in such houses.

Parallel to the other measurements in Ede we therefore sampled volatile hydrocarbons on charcoal tubes with a flow of 100 ml/min. Samples were taken in the living room at breathing height over approximately one week.

Simultaneously, ambient air samples were collected on three sites with a varying proximity of traffic. All samples were analysed on constituents in the boiling-point range of 70°C - 270°C by high resolution capillary gas chromatography and flame ionisation detection. Several samples were also analysed by capillary gas chromatography/quadrupole mass spectrometry.

The results reported here concern the occurrence of hydrocarbons and chlorinated hydrocarbons in living rooms in relation to the outdoor concentration.

The outdoor concentrations at the three sites were comparable within 40%, which was considered sufficient to treat the average ambient air pollution within the area of investigation as homogeneous. The distribution of the measured concentration of 45 components together with the indoor/outdoor ratio is shown in Table 6.

The straight chain saturated hydrocarbons, components of paint, varnish and ink, were dominant constituents. The indoor concentration of all measured straight chain hydrocarbons was higher than outdoors expecially from C_9 on (ratios of more than 10).

The elevation was typical for the vast majority of the investigated living rooms. A similar pattern was found for branched chain saturated hydrocarbons. Another class of saturated hydrocarbons are the alicyclics of which the cyclohexanes were the most abundant. Their concentrations were also higher indoors.

Important representants of unsaturated non-aromatic cyclic constituents are the terpenes, components of plant origin. Limonene was generally found in the living rooms in concentrations approximately 50 times higher than outdoors. Apart from limonene also

TABLE 6. Distribution of the hydrocarbon and chlorinated hydrocarbon concentrations ($\mu g/m^3$) over 133 living rooms and the mean indoor/mean outdoor ratio. Indoor air pollution survey Ede, winter 81/82.

	50-pct	90-pct	95-pct	max.	mean indoor	mean outdoor	indoor/ outdoor Ratio
n-hexane	4.2	11	16	107	7.3	3.9	2
n-heptane	2.7	11	20	68	5.3	1.9	3
n-octane	2.2	9.4	26	60	5.2	0.8	7
n-nonane	4.4	32	83	270	18	1.1	16
n-decane	9.5	58	150	430	31	1.6	19
n-undecane	5.2	30	57	190	13	0.8	16
n-dodecane	1.6	9.2	12	120	4.5	<0.3	>15
n-tridecane	1.2	4.4	5.8	19	1.9	<0.3	> 6
n-tetradecane	2.0	3.5	4.4	8.0	2.1	<0.3	> 7
n-pentadecane	1.5	2.7	2.9	3.6	1.5	<0.3	> 5
n-hexadecane	<0.3	1.0	1.6	2.9	<0.3	<0.3	-
3-methylpentane	2.8	7.5	13	100	4.9	2.5	2
2-methylhexane	2.4	8.6	14	54	4.3	2.1	2
3-methylhexane	2.0	6.8	11	44	3.4	1.8	2
cyclohexane	1.1	4.6	6.3	22	2.0	0.5	4
methylcyclohexane	1.5	4.7	9.8	50	2.9	1.0	3
dimethylcyclopentane isomer	<0.3	1.1	1.6	2.8	0.3	<0.3	>1
dimethylcyclopentane isomer	<0.3	0.8	1.3	2.3	<0.3	<0.3	-
dimethylcyclopentane isomer	0.7	2.1	3.7	7.8	1.0	0.5	2
limonene	26	103	138	216	38	0.7	54
benzene	7.1	16	26	150	9.9	6.7	1
toluene	40	98	160	700	55	16	3
xylenes	12	40	79	180	21	10	2
ethylbenzene	3.0	8.8	21	45	5.0	2.9	2
n-propylbenzene	1.0	3.3	5.0	27	1.8	0.5	4
i-propylbenzene	0.3	1.3	3.5	11	0.7	<0.3	>2
o-methylethylbenzene	1.8	8.7	14	72	4.4	0.9	5
m-methylethylbenzene	3.5	15	32	165	8.1	2.1	4
p-methylethylbenzene	1.6	7.4	16	77	4.0	0.9	4
1,2,3-trimethylbenzene	1.4	4.5	6.5	40	2.3	0.5	5
1,2,4-trimethylbenzene	6.4	25	54	280	14	0.6	23
1,3,5-trimethylbenzene	1.5	5.4	13	99	3.6	0.8	5
n-butylbenzene	0.9	5.6	8.7	40	2.3	<0.3	> 8
p-methyl-i-butylbenzene	0.7	2.8	5.5	32	1.6	<0.3	> 5
naphtalene	<0.3	2.6	3.7	14	1.0	<0.3	> 3
1-methylnaphtalene	<0.3	0.6	0.8	2.2	<0.3	<0.3	-
tetrachloromethane	<4	<4	<4	<4	<4	<4	-
trichloroethene	<1.5	<1.5	<1.5	106	<1.5	<1.5	-
tetrachloroethene	<2	5.6	6.8	205	2.5	3.5	1
chlorobenzene	<0.4	<0.4	<0.4	<0.4	<0.4	<0.4	-
m-dichlorobenzene	<0.6	<0.6	<0.6	9.1	<0.6	<0.6	-
p-dichlorobenzene	1.8	14	25	140	7.2	<0.6	>12
1,2,3-trichlorobenzene	<0.8	<0.8	<0.8	2.7	<0.8	<0.8	-
1,2,4-trichlorobenzene	<0.8	<0.8	<0.8	15	<0.8	<0.8	-
1,3,5-trichlorobenzene	<0.8	<0.8	<0.8	8.3	<0.8	<0.8	-

terpenes like α-pinene, β-pinene and to a lesser extent sabinene were detected, but because of coelution of saturated hydrocarbons their concentrations could not be quantified reliably.

Aromatic hydrocarbons are widely used as solvents. Nowadays the benzene content of most solvents is quite small. This possibly is the reason for the comparable benzene pollution indoors and outdoors and the three times higher concentration of toluene, the main constituent of many aromatic solvents. All other investigated alkylated aromatics had indoor concentrations several times higher than outdoors.

The generally occurring chlorinated hydrocarbon in the $\mu g/m^3$ range was p-dichlorobenzene, which is applied as air refresher. The indoor outdoor ratio was at least 12. The solvents trichloroethene and tetrachloroethane were only found occacionally in elevated concentrations. The same applies to the trichlorobenzenes.

Almost all hydrocarbons investigated and at least one of the chlorinated hydrocarbons appeared in much higher concentrations indoors than outdoors. This justifies further investigation to explore the influence of smoking, consumer products, building materials and ventilation on the concentration level of volatile organics.

CONCLUDING REMARKS

In summary, air pollution inside Dutch houses was often much higher than the common outdoor levels in the Netherlands. In several houses existing or proposed standards for ambient air were exceeded. To what extent the health of specific population groups is influenced by indoor pollution deserves further study.

Presently we are conducting two pilot studies concerning the effects of indoor NO_2 on respiratory health. In one of these, personal exposures of a limited number of participants are also measured. Preliminary results of these studies will be presented at the conference.

On the basis of the results obtained so far, we do have the impression that indoor air quality in several cases needs to be improved. Improvement of indoor air quality should preferably start at the source. For some localized sources such as gas appliances, local exhaust systems can be applied.

For RSP and to a lesser extent for CO, the role of the occupants in maintaining indoor air quality is quite clear, since smoking and maintenance of gas appliances are important factors determining indoor concentration levels.

For NO_2 and volatile hydrocarbons the situation is different. For hydrocarbons we still have to identify the major factors governing the concentration levels.
As far as NO_2, installing range hoods and geiser flues in existing houses to reduce NO_2 levels is advisable but often difficult to realise because of the construction of the buildings.

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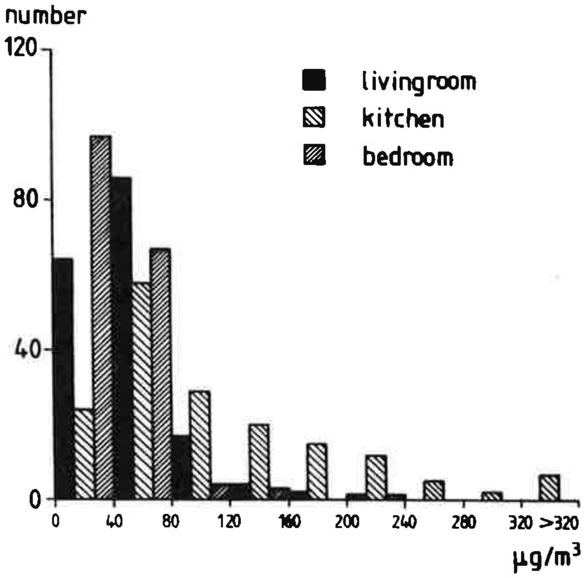

FIGURE 1. Frequency distribution of weekly average NO_2 concentrations in kitchens, living rooms and bedrooms. Indoor air pollution survey Ede, winter 81/82.

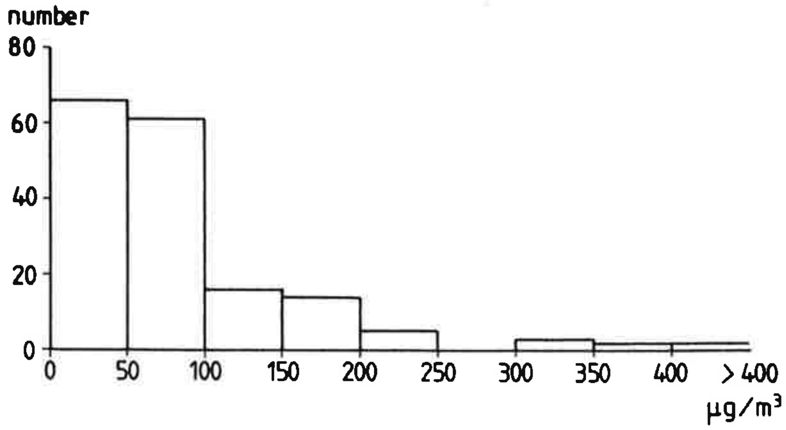

FIGURE 2. Frequency distribution of weekly average concentrations of respirable suspended particulate matter in living rooms. Indoor air pollution survey Ede, winter 1981-1982.