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INDOOR CARBON MONOXIDE AND NITROGEN DIOXIDE POLLUTION IN THE NETHERLANDS

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Introduction

Research on communal air pollution in the past has focussed mainly on the outdoor environment, though most people in Western society spend far more time indoors.
Indoor air pollution is relevant for interpretation of past epidemiological studies and design of future studies on effects of air pollution on health. Indoor air pollution is also important in energy conservation strategies that might restrict indoor - outdoor air exchanges.
Combustion in gas-fired appliances is one of the sources of air pollution indoors. In the mid-sixties a large amount of natural gas became available in the Netherlands, which resulted in a rapid increase in the use of domestic gas appliances in Dutch households. In 1978 over 90% of the houses was connected to the gas network and about 70% had a "geiser", a flow-through type of water heater, usually located in the kitchen and in most cases unvented. Nevertheless little systematic information is available on levels of indoor air pollution from gas-fired appliances.

In this connection we started a study on indoor air pollution, the major components of interest being carbon monoxide, nitrogen dioxide, respirable suspended particles and sulfur dioxide. For the winter of 1981 - 1982 a program has been set up for continuous short-time measurements in about 15 houses, and for weekly averaging measurements in about 200 houses.
In addition to air pollution we will also measure temperature, humidity, ventilation of the kitchen and transfer of gases from kitchen to living-room and bedroom with a tracer technique.
As part of this project, a survey was carried out in the fall of 1980 to levels of carbon monoxide and nitrogen dioxide from gas-fired cooking and water-heating appliances in the cities of Arnhem and Enschede.

As geisers are notorious for their sometimes high production of carbon monoxide, the houses were selected on the presence of a geiser. Levels of more than four percent carbon monoxide by volume in the flue gases have been reported in extreme cases (Bartholomeus et al., 1976).
Less is known about the production of nitrogen dioxide. The two cities were chosen because of a difference in maintenance system. In Enschede a central maintenance firm services about 80% of the geisers each year by replacing the burner for an ultrasonic cleaned exchange burner.
In Arnhem less regularly only on-site cleaning is carried out by different firms. For Enschede also earlier data (1968) exist for CO production from geisers and resulting levels in kitchens (Borst, 1972).

Methods and Materials

In Enschede and Arnhem a random sample of houses with geisers of 278 and 537 respectively was drawn. In Enschede the sample was stratified toward houses not participating in the local maintenance system, otherwise too few houses in this category would be available for statistical analysis. Later in a number of houses a geiser appeared not to be present. In table 1 the participation to the survey is given. The houses were alternately visited in Arnhem and Enschede from October 27 to December 19, 1980. In each house a number of variables was assessed in relation to CO, of which the most important are given in figure 1.

Measurements in the kitchen were performed with closed doors and windows. Permanent ventilation-ducts were not closed. The following measurements were carried out :

- CO (Ecolyser 2000) and CO_2 (Draeger tubes) at breathing height before and after continuous operation of the geiser for 15 minutes.
- CO in the flue gas after 1 and 15 minutes of continuous operation of the geiser.
- CO concentration increase 0.2 m under the ceiling continuously during 15 minutes of operation of the geiser.

After the measuring period the interference-filter for NO in one of the two monitors appeared to be saturated by the extreme humidity in the flue gases. The measurements with that monitor were not included in further statistical analysis, so that 254 sets of reliable data were available (177 for Arnhem and 77 for Enschede).

House characteristics of the resulting sample were not significantly different from the original sample.

Measurements of NO_2 with Palmes diffusion tubes (Palmes, 1976) were done in the kitchen and living-room at breathing height in the same houses as the CO measurements. The tubes were placed as far away as possible from gas appliances, hood, ventilation-ducts, windows and doors. After 5 to 8 days the tubes were collected and analysed in our laboratory. Unlike the CO data, which are collected under standardized conditions, the NO_2 measurements represent normal living conditions.
In addition to the measurements an inquiry was held with a questionnaire to the housewive. The questions were dealing with complaints in relation to CO exposure, use of geiser and other gas appliances and personal and house characteristics.

Results and Discussion

The main results of the CO measurements are given in table 2.
A CO concentration of 300 ppm in the flue gases is the level above which local gas companies usually require cleaning and adjustment. In Arnhem 31% and in Enschede 16% of the geisers did not meet this requirement. In 20% of the kitchens in Arnhem and 11% in Enschede, a CO level of more than 50 ppm was reached at breathing height after 15 minutes of operation of the geiser. The installation codes for gas appliances require a.o. that the CO concentration in the house shall not exceed 50 ppm during normal operation.

CO in flue gases

In table 3 the statistical relation is given between CO concentration in flue gases after 1 minute of operation of the geiser and a number of other variables.

A strong correlation was found with the presence of an atmospheric safety provision, which turns the geiser of if the CO_2/O_2 ratio becomes too high. Also a strong correlation was found with the burner type. Diffusion burners give less CO than Bunsen burners. As both variables are not independent, all geisers with diffusion burners have an atmospheric safety provision, on statistical grounds no conclusion can be drawn which factor is most important. It seems likely, however, that burner type is responsible for the correlation, because Bunsen burners can get easier dirty by premixing with air. The weak correlation with make of the geiser appeared to depend also on burner type, as no correlation was found if only geisers with Bunsen burners were taken into account. No or only a weak correlation was found with maintenance system and city. However, when only is looked at geisers with a Bunsen burner these correlations become significant in the way that Arnhem scores higher CO values in the flue gases. This absence of a correlation for all geisers is due to the presence of more geisers with diffusion burners in Arnhem.

The increase of CO concentrations in flue gases from 1 to 15 minutes of operation of the geiser was only found to be significantly correlated to the CO_2 concentration in the kitchen after 15 minutes and the presence of a flue. The mutual dependance of these variables is obvious.

Using the statistical technique of linear logit models, it was shown that burner type and maintenance system were the main factors influencing the CO levels in the flue gases.

CO in kitchens

The statistical relation was investigated between CO increase under the ceiling and at breathing height and other variables. The variables presence of a flue, CO_2 concentration after 15 minutes and atmospheric safety provision appeared to be correlated significantly with CO increase. Also the difference between Arnhem and Enschede was significant. In Enschede less often a concentration was found of more than 100 ppm under the ceiling (11.8% of the cases) and at breathing height (2.7% of the cases) than in Arnhem (25.7% and 8.7% of the cases respectively). However, in Arnhem more geisers were found with a diffusion burner and in Enschede more geisers with a flue. The combined effect of these parameters was studied with linear logit models. It was shown that the main independant factors influencing the CO increase were burner type and presence of a flue. Apart from these, a difference remained between Arnhem and Enschede that could not be attributed to one of the other assessed variables.

Relation between observed CO concentrations and reported complaints

In the questionnaire a number of questions was asked about possible complaints from CO exposure. Statistical analysis of these complaints in relation to the observed CO concentrations at breathing height did not reveal any association. We must consider, however, that symptoms of CO only can be found above the level of 3% of carboxyhemoglobin. To reach this level concentrations much higher than 25 ppm are necessary As too few concentrations at these levels were found it could hardly be expected to find a relationship. Another factor is that only a bad estimate of the actual exposure to CO can be derived from our data.

NO_2 measurements

The results of the NO_2 measurements are compilated in table 4. The frequency distribution is given in figure 2.

The 30 houses with a ratio higher than one had, with two exceptions, differences between kitchen and living room of only a few ppb. Outdoor concentrations of NO_2 varied from 20 - 50 $\mu g/m^3$ during the same period. The NO_2 concentration in the kitchen was greatly influenced by the presence of a geiser-flue and by the presence of a ventilation-hood above the cooking range. After allowing for the presence of a flue and a ventilation-hood, associations were found with type of space-speating, use of shower and socioeconomic status of the occupants. Central heating systems had lower NO_2 concentration, while in houses with shower use continuously over longer periods and in houses with people of lower socioeconomic status higher NO_2 concentrations were found.

In the living room the NO_2 concentrations were also influenced by the presence of a geiser-flue. Using linear logit models with the NO_2 concentration as response variable, a good fit was found for models with the NO_2 concentration in the kitchen and the number of doors and rooms between kitchen and living room as independant variables. The models were significantly improved when type of heating or number of occupants were added. Higher NO_2 concentrations were found in houses with gas-fired zone-heating and in houses with four or more occupants.

Conclusions

In test requirements for geisers a limit of 600 ppm CO in the flue gases is set for safe use. In 18% of the cases this requirement was not met. Local gas companies often use 300 ppm as a limit. This concentrations was exceeded in 27% of the cases. This study showed that geisers with a diffusion burner had a lower chance of exceeding the limit. Only one make has such a construction. For geisers with Bunsen burners the maintenance system had a great influence. The system applied in Enschede with off-site ultrasonic cleaning appeared to be superior. This does not mean that geisers with a diffusion burner do not have to be cleaned.
The Dutch installation code requirement of 50 ppm, for which no averaging time is given, was exceeded in 17% of the cases.
No health-based indoor air quality standards exist for CO. However, indoor concentrations can be compared to outdoor air quality standards, as the same people to be protected are involved. The proposed Dutch outdoor standard reads (Gezondheidsraad, 1975) : 35 ppm 1 hour average, not to be exceeded more than once a week ; 9 ppm 8 hour average, not to be exceeded more than once a month. Although our measurements were done under standardized conditions, it is obvious that this standard is frequently violated indoors.

In addition to the burner type and the maintenance system the presence of a flue was the main factor in the CO concentration at breathing height.
Unlike CO- NO_2 measurements were done under normal living conditions. For outdoors the Dutch Health Council has proposed a standard for NO_2 (Gezondheidsraad, 1979), which reads as follows : 135 $\mu g/m^3$ (av. for 1 h.) not to be exceeded during 98% of the time, 300 $\mu g/m^3$ not to be exceeded more than once a year ; 120 $\mu g/m^3$ (av. for 24 h.) not to be exceeded during 98% of the year, 150 $\mu g/m^3$ not to be exceeded more than once a year.
Short term exposures to high levels of NO_2 decrease the resistance to airway infections and cause irritation of the mucus membranes of the respiratory system. The non-continuous use of unvented gas appliances produces high peak concentrations during use. Peak-to-mean ratios of 5 to 10 have been reported (Apling et al., 1979). This is also illustrated by own measurements in a flat (figure 3).

Although averaging times in our study are different from those in the standard, it is obvious that the outdoor standard will regularly be exceeded by indoor concentrations.
Higher NO_2 concentrations were found in kitchens and living rooms of houses with an unvented geiser and in houses with gas-fired zone-heating. Elevated NO_2 levels in the living room were associated with the absence of doors and rooms between kitchen en living room and with a higher number of occupants. Long use of the shower and the absence of a ventilation-hood were also found to explain statistically higher concentrations in the kitchen.

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TABLE 1 PARTICIPATION TO THE INDOOR AIR POLLUTION SURVEY OF CO AND NO_2 IN ARNHEM AND ENSCHEDE, 1980

	ENSCHEDE		ARNHEM	
	NUMBER	%	NUMBER	%
PARTICIPATING	112	40.3	237	44.1
NOT AT HOME (2x)	65	23.4	127	23.6
NO GEISER	49	17.6	49	9.1
EMPTY HOUSE/MOVED	11	4.0	17	3.2
REFUSAL	34	12.2	104	19.4
VARIOUS	7	2.5	3	0.6
	278	100.0	537	100.0

TABLE 2 CO LEVELS UNDER STANDARDIZED CONDITIONS IN FLUE GASES OF GEISERS AND IN KITCHENS. INDOOR AIR POLLUTION SURVEY ARNHEM AND ENSCHEDE, 1980

CO LEVELS IN FLUE GASES AFTER 1 MIN. OF OPERATION (PPM)			CO LEVELS AT BREATHING HEIGHT AFTER 15 MIN. OF OPERATION (PPM)		
	N	%		N	%
< 100	119	53	< 10	154	63
101-300	46	20	11-50	50	20
301-600	20	9	51-100	25	10
> 600	41	18	> 100	17	7
MISSING	28			8	
	254	100		254	100

TABLE 3 STATISTICAL RELATION BETWEEN CO CONCENTRATION IN FLUE GASES AFTER 1 MIN. OF OPERATION OF THE GEISER AND OTHER VARIABLES. INDOOR AIR POLLUTION SURVEY ARNHEM AND ENSCHEDE, 1980

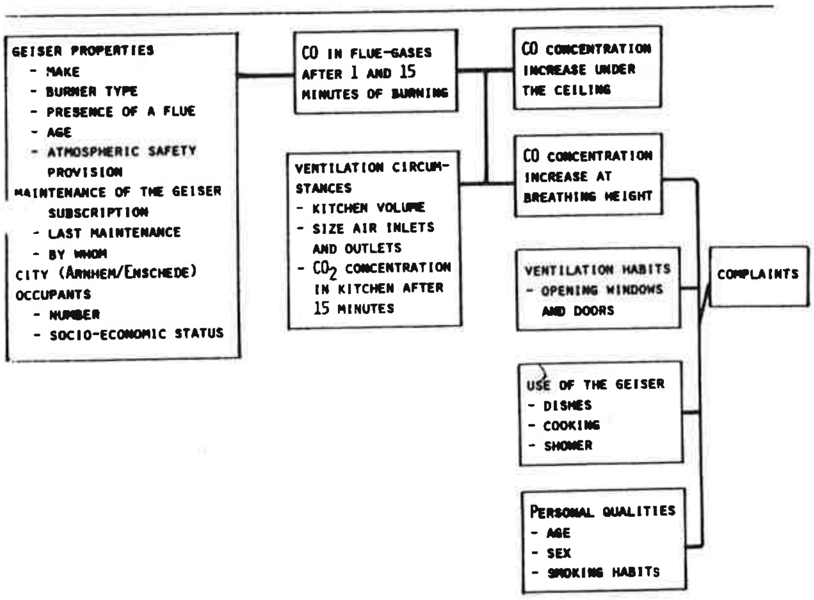
VARIABLES	SIFNIFICANCE 1) (P)	NUMBER
MAKE OF THE GEISER	.07	219
PRESENCE OF A FLUE	N.S. 2)	223
ATMOSPHERIC SAFETY PROVISION	< .01	201
BURNER TYPE	< .01	219
VOLUME OF THE KITCHEN	N.S.	223
AGE OF THE GEISER	N.S.	162
MAINTENANCE SYSTEM	N.S.	131
CO_2 CONC. IN KITCHEN AFTER 15 MIN.	N.S.	226
SOCIOECONOMIC STATUS	N.S.	192
ARNHEM/ENSCHEDE	.09	226

1) CHI-SQUARE TEST
2) NOT SIGNIFICANT $P \geq 0.1$

TABLE 4 NO_2 CONCENTRATIONS IN KITCHENS AND LIVING-ROOMS AND RATIOS BETWEEN LIVING-ROOM AND KITCHEN. INDOOR AIR POLLUTION SURVEY ARNHEM AND ENSCHEDE, 1980

	ARITH. MEAN IN $\mu G/M^3$ (PPB)	RANGE
NO_2 CONC. IN KITCHENS (N = 286)	118 (63)	35 - 472 (20 - 251)
NO_2 CONC. IN LIVING-ROOMS (N = 291)	58 (31)	35 - 346 (20 - 184)
RATIO LIVING-ROOM / KITCHEN	0.61	0.08 - 3.34

FIGURE 1 VARIABLES ASSESSED IN RELATION TO CO. INDOOR AIR POLLUTION SURVEY IN ARNHEM AND ENSCHEDE, 1980



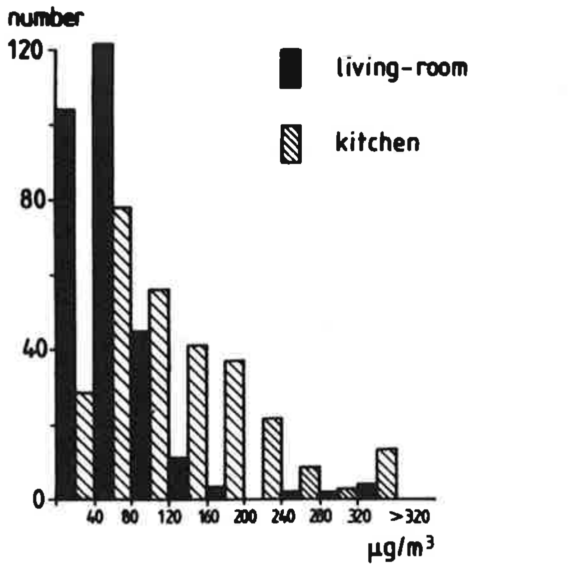


FIGURE 2 FREQUENCY DISTRIBUTION OF NO_2 CONCENTRATIONS IN 289 KITCHENS AND 294 LIVING-ROOMS. INDOOR AIR POLLUTION SURVEY ARNHEM AND ENSCHEDE, 1980