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INDOOR NO_x POLLUTION

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INTRODUCTION

Over the past two decades a considerable amount of money has been spent worldwide at attainment of a healthier ambient air quality. Almost all of this effort was directed to the outdoor environment. The Dutch law on air pollution for example restricts itself explicitly to the outdoor environment in its definition of air pollution.

Modern man, however, spends on the average 90% of his time indoors. Biersteker et al. [1,2] were the first to pay attention to the role of indoor pollution in air pollution epidemiology. Their work focussed on indoor-outdoor relationships for SO_2 and smoke in 60 Rotterdam homes. They showed that part of the increases in mortality and morbidity during air pollution episodes may be caused by indoor production of CO, SO_2 and smoke. Due to the use of cleaner natural gas for home heating in the Netherlands since 1965, it was assumed that the problem of indoor air pollution was automatically solved.

Only recently indoor air pollution is getting increasing attention again thanks to energy conservation strategies that may restrict indoor-outdoor air exchange. In that context an increasing amount of evidence has shown that concentrations of some pollutants indoors frequently exceed those outdoors, particularly for those pollutants which can also be generated indoors like the oxides of nitrogen [3].

Combustion of gas in gas-appliances at high temperatures always generates oxides of nitrogen from nitrogen and oxygen in the air. The amount generated and the ratio in which NO and NO_2 are formed varies with burner construction and condition [4, 5, 6]. From the health point of view NO_2 is more important than NO, which is far less toxic [7] . Although indoor NO concentrations are found even higher than NO_2 concentrations, NO does not play an important role indoors, as residence times are too short to allow for oxidation to NO_2 [5, 8].

In a number of studies higher NO_2 levels are established inside homes that cook with gas compared to homes with electric stoves [9, 10, 11, 12, 13]. The impact of smoking on indoor NO_2 levels is small. Good et al. [14] found values of 22 and 19 $\mu g/m^3$, respectively in smoking and nonsmoking households in the winter in houses without gas stoves.

In our contribution indoor emissions from gas stoves, ovens and water-heaters will be discussed. Also the results of a large survey of NO_2 levels in Dutch homes with a geiser, a flow-though type of waterheater, will be presented in addition to the preliminary results of a more general survey. The potential impact on health will be discussed and finally some comments will be made about how to cope with the problem of indoor air pollution, as it is obvious that problems exist to an extent that action is desirable by policymakers.

NO_2 EMISSIONS FROM GAS-APPLIANCES

In several studies NO_2 emissions from domestic gas-appliances have been measured [15, 16, 17, 18]. For ovens emissions were found of 3 - 7 cm^3 NO_2/MJ and for cooking-burners of 4.5 - 10 cm^3 NO_2/MJ. In the Netherlands NO_x emissions were measured of 7 - 10 cm^3 NO_x/MJ for water-heaters and 6 cm^3 NO_x/MJ for an oven [19, 20]. Using a NO/NO_2 ration of 3 for waterheaters and 1 for the oven, according to our own observations, these NO_x emissions can be converted to NO_2 emissions of 2 cm^3/MJ for waterheaters and 3 cm^3/MJ for the oven.

In terms of exposure these figures as such are meaningless. However, if these emissions are compared with CO_2 emissions, a ratio can be calculated of 1 - 4 mg NO_2 on 10 g CO_2. In kitchens a CO_2 concentration is allowed of 0.5% CO_2 according to the Dutch installation code, which corresponds to a NO_2 concentration of about 0.9 - 3.6 mg/m^3. The concentration we actually found in Dutch homes will be presented in the next paragraphs.

NO_2 IN DUTCH HOMES

To measure NO_2, passive diffusion samplers developed by Palmes were used [21]. These are small, 8 cm long, 1 cm inner diameter acrylic tubes with stainless stell wire mesh, coated with triethanolamine, inserted in one end. When open to the atmosphere with the other end, NO_2 molecules diffuse at a rate proportional to the ambient concentration. The absorbed NO_2 is, after dissolving, analyzed spectrophotometrically. These devices have been compared with standard chemiluminescent NO_2 monitors by Warren Spring Laboratory [22]. Accuracy has been demonstrated to be better than $\pm$ 10%. Replicate samples indicated a coefficient of variation of less than 10%. A lower detection limit of about 600 ppb x hours makes these devices very suitable for integrated measurements of NO_2 levels for week long periods.

The measurements were done in a random sample of about 300 houses with a geiser in Enschede and Arnhem. Geisers are present in about 70% of the Dutch homes. This study was mainly designed to assess the influence of the geiser on CO levels and possibly related health complaints. The CO results are published elsewhere [23]. The houses were alternately visited in Arnhem and Enschede from October 27 to December 19, 1980. In each house NO_2 tubes were placed in the kitchen and living-room at breathing height 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. In addition the housewives were interviewed about frequency and duration of use of the geiser and other gas-appliances, and about personal and house characteristics.

The results of the NO_2 measurements are given in table 1, while the frequency distribution is given in figure 1.

TABLE 1
Weekly average NO_2 concentrations in kitchens and living-rooms
ratios between living-room and kitchen
Indoor air pollution survey Arnhem Enschede, 1980

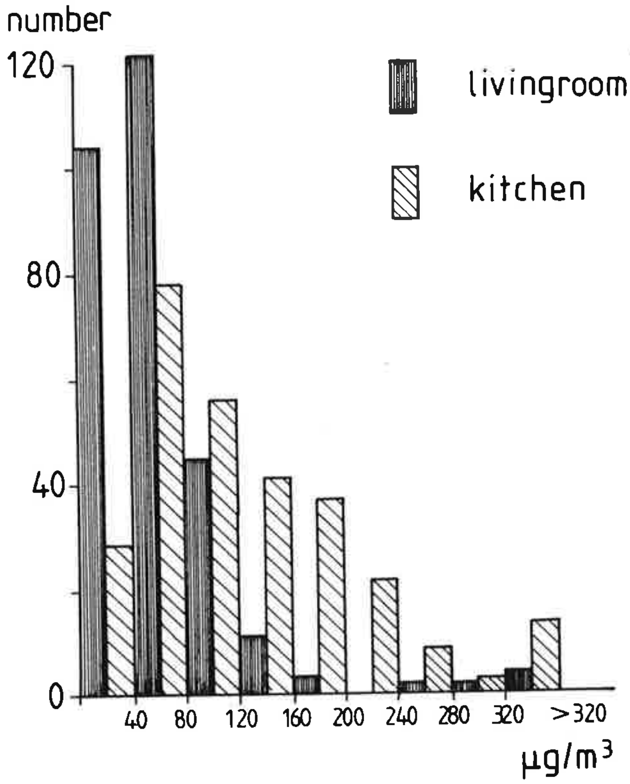
	Arithm. mean in µg/m^3 (ppb)	range
NO_2 conc. in kitchens (n = 286)	118 (63)	35 - 472 (20 - 251)
NO_2 conc. in living-rooms (n = 291)	58 (31)	35 - 346 (20 - 184)
Ratio living-room/kitchen	0.61	0.08 - 3.34

Weekly average outdoor concentrations of NO_2 at the nearest stations of the Dutch air pollution measuring network during the same period varied from 20 - 50 µg/m^3.

Statistical analysis of the results showed that the presence of a flue on the geiser and the presence of a ventilation hood above the cooking range had the greatest influence on the NO_2 concentrations in the kitchen. After allowing for the presence of a flue and a ventilation hood, associations were found with type of space-heating, use of shower and socioeconomic status of the occupants. Houses with central heating systems had lower NO_2 concentrations, while in houses with prolonged shower use and in houses with people of lower social class higher NO_2 concentrations were found.

In the living-room NO_2 concentrations were also influenced by the presence of a flue on the feiser. Using linear logit models with NO_2 concentration as respons variable, a good fit was found for models with "NO_2 concentration in kitchen" and "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 central heating systems and in houses with four or more occupants.

FIGURE 1
Frequency distribution of weekly average NO_2 concentrations
in 289 kitchens and 294 living-rooms
Indoor air pollution survey Arnhem and Enschede, 1980



A second survey was held during the winter of 1981/1982. A sample of 326 houses in the town of Ede, built after world war II, was randomly chosen from the municipal housing registration. Duplicate NO_2 samples with averaging times of 4 - 6 days were taken in the kitchen, bedroom and living-room of 175 houses, 89 occupants (27%) refused, 62 occupants (19%) were found not at home twice or more. In addition to NO_2 other parameters were also assessed in this survey like carbon monoxide, respirable suspended particles, organics, temperature, relative humidity, ventilation of the kitchen, transfer of gases from kitchen to living-room with a tracer technique, and personal and house characteristics.

In table 2 the results of the NO_2 measurements are given. Figure 2 gives the frequency distribution.

Weekly average outdoor concentrations of NO_2 at the nearest stations of the Dutch air pollution measuring network during the same period varied from 25 - 72 $\mu g/m^3$ with a mean of 44 $\mu g/m^3$.
Further statistical analysis of the results in relation to controlling factors is in progress.

TABLE 2

Weekly average NO_2 concentrations in kitchens, living-rooms and bedrooms in Dutch homes.

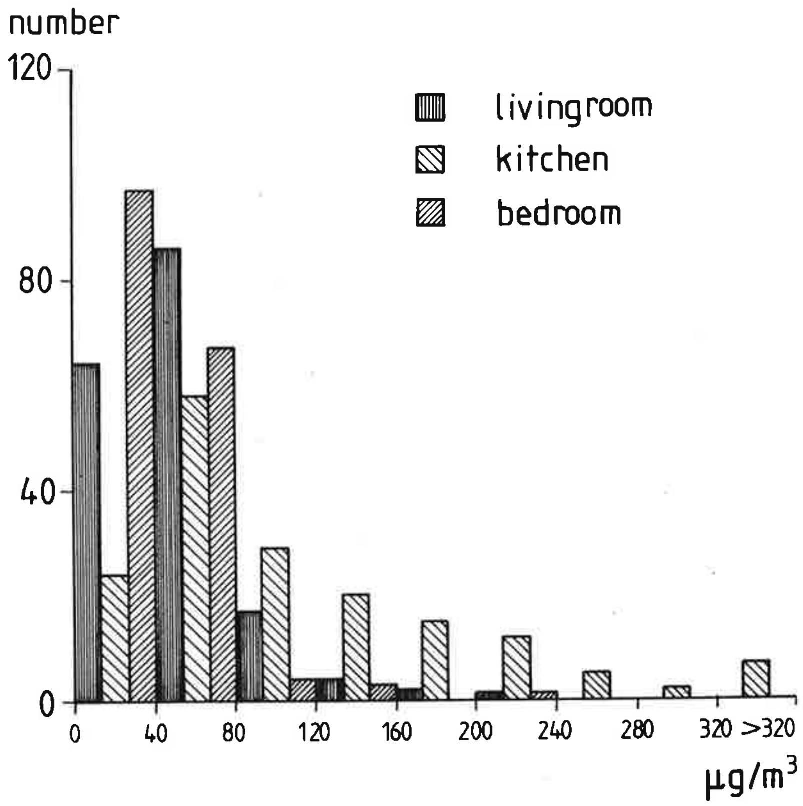
Indoor air pollution survey Ede, winter 1981/1982

		Arithm. mean in µg/m³ (ppb)	Range
NO_2 conc. in kitchens	(n = 173)	115 (61)	11 - 721 (6 - 384)
NO_2 conc. in living-rooms	(n = 174)	54 (29)	11 - 222 (6 - 118)
NO_2 conc. in bedrooms	(n = 172)	43 (23)	11 - 201 (6 - 107)

FIGURE 2

Frequency distribution of weekly average NO_2 concentrations in 175 Dutch homes

Indoor air pollution survey Ede, winter 1981 - 1982



DISCUSSION

No health based indoor air quality standards exist for NO_2, but indoor concentrations can be compared to outdoor air quality standards, since the same people to be protected are involved.

For outdoor air the Dutch Health Council has proposed a standard which is given in table 3 [24].

TABLE 3
Outdoor air quality standard proposed by the Dutch Health Council [24].

Percentile	Averaging time	
	1 h.	24 h.
95	110	100
98	135	120
99.7 (1 day/year)	-	150
99.99 (1 hour/year)	300	-

Concentrations in $\mu g/m^3$

This standard is in line with the international tendency to regulate particularly short term exposures [7]. Short term exposures to high levels of NO_2 decrease the resistance to airway infections and may have bronchoconstrictive effects on the respiratory system of sensitive persons [7]. Although in our study the averaging time was approximately one week the results can be compared with the 24 h standard, since one week can be considered as 7 daily cycles. Figure 1 and 2 show that it is obvious that the outdoor standard is regularly exceeded indoors. Comparison with the 1 h standard is more difficult. In other studies peak to mean ratios of 5 to 10 have been found in rooms with unvented gas-appliances [22, 25], which indicates a high probability that short term exposures are also in excess of the standard.

From the health point of view there is some recent evidence that gas cooking in homes is associated with an increased incidence of lower respiratory disease in children. Melia et al. [12, 26, 27] examined 1810 British children aged from 5 to 11 years, living in gas-cooking homes, and 3017 living in electric-cooking homes. They found a higher prevalence of respiratory symptoms for children living in gas-cooking homes, but only in urban areas.

In another study [28] with more than 8000 6 to 10 year old children in 6 U.S. cities a greater history of respiratory disease before age 2, as reported by the parents, and small but significant differences in pulmonary function were associated with the presence of gas stoves in the homes. The consistency of this finding across each of the 6 cities is interesting.

Hasselblad et al. [29] also found a small decrease in forced vital capacity among girls, associated with the presence of a gas stove in the home, in a study of over 16,000 children. These studies are in contrast with that of Keller et al. [30] who found no differences in respiratory symptoms among 600 school-aged children on the basis of type of stove. However, the small sample size and methodological shortcomings make it unlikely that small effects as reported above could have been detected. It has been generally assumed that NO_2 exposure is responsible for the health effects associated with gas-cooking. However, both Florey et al. [27] and Letz et al. [31] were unable to find correlations between NO_2 levels in kitchen and living-room and respiratory functions. The former group found instead a week association between NO_2 in bedroom and respiratory functions and illness.

Another feature of indoor NO_2 pollution is the important role it plays in the interpretation of past epidemiological studies and design of future studies on effects of air pollution. In the past in most cases only outdoor pollution was used as independent variable. Indoor pollution, however, can be the real cause of some of the associations that have been found leading to an overestimate of health hazards of outdoor pollution when only outdoor levels are measured. In a recent study Dockerey et al. [32] compared indoor, outdoor and personal exposure to NO_2. Members of families cooking on gas were found to have exposures twice as high as in families cooking electric and twice as high as the outdoor levels. They concluded that the differential NO_2 exposures in homes with and without gas stoves cannot be left out in epidemiologic studies of the health effects of air pollution.

Finally we would like to comment on how to deal with indoor pollution in homes. In many countries minimum ventilation requirements exist on the basis of CO_2 and odor production by the occupants. These ventilation requirements are derived from the early work of Yaglou et al. [33, 34]. Energy conservation urges to decrease ventilation, which gives rise to potential accumulation of indoor generated pollutants. On the other hand ventilation has been often suggested and applied as a tool to decrease concentrations of hazardous pollutants. In our opinion ventilation may only be applied to keep concentrations within certain limits for strictly inevitable pollutants. Only CO_2, odors and humidity produced by the occupants can be considered as such. All other pollutants have to be reduced at the source or even by complete banning of the source. In some cases local exhaust can be applied if emissions indoors occur locally, which is the case for emissions from gas-appliances.

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