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CONTINUOUS AND PASSIVE MONITORING OF NITROGEN DIOXIDE IN UK HOMES

DAVID ROSS

Building Research Establishment, Watford, Herts, WD2 7JR, United Kingdom

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ABSTRACT

The UK Building Research Establishment has carried out measurements of levels of nitrogen dioxide (NO_2) in 12 homes in the South of England. Two types of detection device were used: the Scintrex LMA-3 continuous NO_2 analyser and the Palmes passive diffusion tube. NO_2 concentrations were recorded using both devices in the kitchen, living room and a bedroom of each home for a period of a week. Gas cooking was observed to be the most important source of peak concentrations of NO_2 in the home. Six homes, out of the ten that used gas for cooking, had levels of NO_2 which exceeded the WHO 1-hr guideline value of 210ppb. In at least two of the homes, personal exposure is expected to have also approached or exceeded this guideline value. There was a fairly good correlation of measured average levels of NO_2 between the two detection devices. There was also a fairly good correlation between the 7-day average levels recorded by the diffusion tubes and the maximum 1-hour averages recorded by the continuous detectors. This offers the possibility of gaining information on short-term exposure levels from long-term diffusion tube data. An investigation of the effect of a cooker hood on NO_2 levels in a kitchen suggested that it could prove to be an effective means of removing NO_2 generated during cooking, especially if there are no other competing means of ventilation such as open doors.

Keywords: Nitrogen dioxide, indoor, continuous-measurement, passive-measurement, cooker hood

INTRODUCTION

Nitrogen dioxide (NO_2) is a major pollutant of outdoor air and has received considerable attention. However a previous UK study has shown that 70-75% of personal exposure to NO_2 actually occurs within the home (1). The indoor level is dependent on the NO_2 in the outdoor air infiltrating into the home. Perhaps more importantly, the indoor level also depends on the use of indoor sources of NO_2; principally unvented or poorly vented combustion appliances, e.g. gas cookers and gas and oil fueled space heaters. Exposure to NO_2 may be associated with both chronic and acute adverse health effects primarily in the lungs. Of particular concern is the effect of NO_2 on children and asthmatics.

A number of studies have been performed to investigate the levels of NO_2 in UK homes. Predominantly, these studies have used passive diffusion tubes to monitor indoor levels of NO_2, typically for 7 to 14-day periods (1-3). Whilst passive sampling provides data on the mean levels of NO_2 during the period of tube exposure, it provides no details on any short-term peak concentrations that may occur. Some animal toxicology studies suggest that these peak levels are of more significance to the toxicity of NO_2 than integrated dose (4,5). Hence, the need for continuous monitoring of NO_2 in UK homes.

Previously, Stevenson (6) performed continuous monitoring of NO_2 in 5 UK homes. Measurements were made in three kitchens, two with gas cookers and one all electric, and two living rooms, one heated by a portable butane heater and the other by a paraffin stove. The data showed that combustion appliances can produce NO_2 levels that exceed ambient air quality guidelines.

To investigate further, the Building Research Establishment (BRE), under contract to the UK Department of the Environment, performed both continuous and passive sampling of NO_2 for one week in the kitchen, living room and bedroom of twelve homes in the South of England. This provided data on both peak and integrated exposure and allowed comparisons to be made between the two sampling techniques. This study also provided information on the transport of NO_2 from the source through the home. The experimental method and results are presented within this paper.

METHODOLOGY

Continuous monitoring of NO_2 was performed using Scintrex LMA-3 analysers (7). These analysers have a non-linear detector response and multi-point calibrations were necessary to characterise this. A linear fit was used for levels below 100ppb whilst for higher levels (100-1000ppb) quadratic fitting was required. The measurements were accurate to approximately 12%. Our NO_2 calibration gases were compared against the NO_2 standard gases used by the DoE Air Quality Monitoring Network.

An LMA-3 device was placed, encased in a sound-proof box, in the kitchen, living room and a bedroom of each home for a week. The sample points were located as close to the centre of the room as possible. The inlets of the sample lines were positioned at a height between 1.5 and 2m to correspond to typical breathing heights and thus allow for any stratification of the NO_2 produced at elevated temperatures during combustion. The sample points were at all times located at least one metre horizontally away from any potential source. Samples were taken every three seconds and recorded as 30 second averages.

In parallel with the continuous monitoring, passive monitoring was undertaken with Palmes diffusion tubes (8). The tubes were located close to the continuous sampling points. Additional tubes were placed at the front and back of each home to determine outdoor NO_2 concentrations.

For a one week sampling period, accuracy has previously been demonstrated to be within ±10% with precision better than 2ppb (9,10). Due to the high level of accuracy reported, single tubes were initially used in each room. However duplicates were subsequently found to be necessary (see Section 3.2.1).

To correlate the NO_2 measurements with the usage of combustion appliances, the occupants filled out three questionnaires:

(I) a main questionnaire completed prior to the experiment in order to provide details of the home, the methods of cooking and heating and means of ventilation.

(ii) a daily diary of combustion appliance usage during the experiment, which included the approximate time and duration for which each appliance was on.

(iii) a short questionnaire after the experiment to assess the types of ventilation used in the house during the course of the experiment.

RESULTS

Continuous Measurement

Figure 1 presents a NO_2 peak recorded with a LMA-3 detector in a kitchen. As this figure clearly shows, the peak corresponds to a period of gas cooker use. The peak comprises a number of peaks of shorter time intervals. This may have been due to (a) the effects on ventilation rates caused by the kitchen window being open, and/or (b) the adjustment of the burner setting during cooking.

A detailed comparison of the recorded data from each home and the completed diaries of cooker usage revealed that whenever a gas cooker was used there was always a clearly observable peak in the kitchen NO_2 levels. Comparable sized peaks were observed from hob, grill and oven use. In most cases, there was also a corresponding peak of smaller magnitude in the living room and bedroom data due to the transport of NO_2 around the house. These latter peaks typically occurred after the kitchen peak and extended over larger time periods.

A summary of the results recorded by the LMA-3 analysers is shown in Table 1. The lowest recorded NO_2 levels occurred in the two houses which used electrical cooking appliances.

In the recorded data, only home K showed peaks not corresponding to gas cooker usage. Elevated levels of 70 - 100ppb were recorded a number of times in the kitchen during the monitoring period. The peaks corresponded to periods when the central heating system was on and must have arisen from the balanced-flue boiler used for heating. Figure

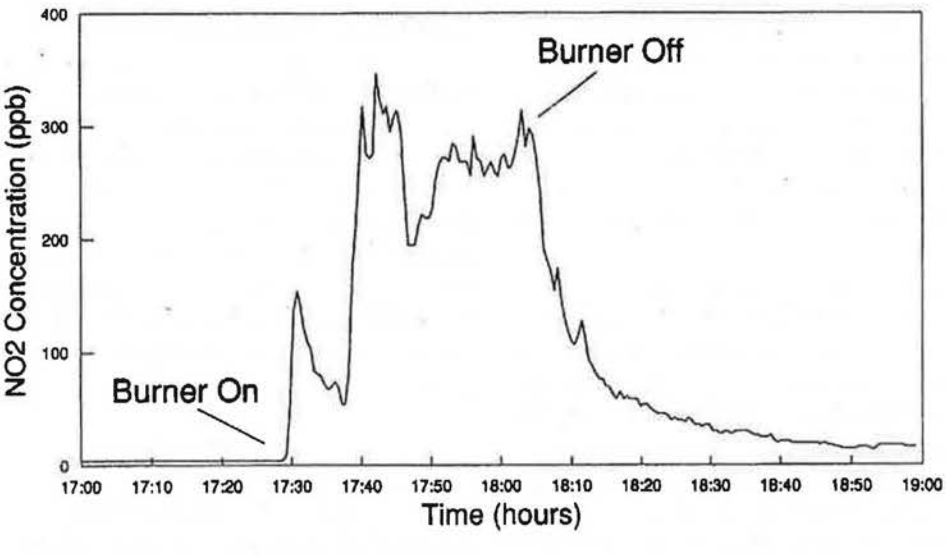

Figure 1. NO2 Levels in a Kitchen

Table 1. Results from the LMA-3 Continuous NO_2 Analyser

House	Start Date	Cooking Fuel	Room	Weekly Average NO2 Level (ppb)	Maximum 1-Hr Average (ppb)
A	25/5/93	Gas	Kitchen	38	215
			Living Room	16	70
B	15/6/93	Gas	Kitchen	22	259
			Living Room	14	120
			Bedroom	18	180
C	22/7/93	Gas	Kitchen	21	282
			Living Room	15	102
			Bedroom	16	115
D	6/8/93	Gas	Kitchen	18	125
			Living Room	15	100
			Bedroom	11	41
E	23/8/93	Gas Hob	Kitchen	13	53
		Electric Grill	Living Room	9	37
		and Oven	Bedroom	4	34
F	21/9/93	Electric	Kitchen	12	44
			Bedroom	8	28
G	5/11/93	Electric	Kitchen	3	14
			Living Room	0	5
			Bedroom	3	19
H	18/11/93	Gas	Kitchen	32	593
			Living Room	25	295
			Bedroom	15	305
I	17/1/94	Gas	Kitchen	22	320
			Living Room	13	191
			Bedroom	10	250
J	7/2/94	Gas	Kitchen	17	100
			Living Room	10	56
			Bedroom	2	16
K	21/2/94	Gas	Kitchen	25	169
			Living Room	11	49
L	12/5/94	Gas Hob	Kitchen	33	211
		Electric Grill	Bedroom	21	115
		and Oven			

2 shows the measured data for one of these peaks. The detailed structure is due to the boiler's thermostat switching the heating system on and off. It is uncertain whether the elevated concentrations arose from leakage of the system or re-entrainment of the combustion gases from outside the home, and this will require further investigation. In this study there were no observable peaks when gas fires (all open-flued) and boilers in other households were used.

It is interesting to observe that in four of the homes which used gas for cooking (B, C, H and I), the maximum levels recorded in the bedroom were higher than those in the living room. Two of these homes (C and I) were flats with all rooms on the same level and hence the results were not unexpected. Homes B and H were two-storey houses, with the kitchen and living room on the ground floor, and the bedroom, where monitoring was undertaken, on the first floor. In both of these cases the internal kitchen door was close to the stairway. It is likely that the buoyancy of the hot combustion gases caused the NO_2 to rise up the stairs which resulted in higher levels in the bedroom than the living room.

The World Health Organisation (WHO) recommends a maximum 1-hour NO_2 air quality guideline level of 210ppb (11). As Table 1 shows, NO_2 levels in the kitchens of six of the houses exceeded this value. Moreover, houses H and I had cooking periods during which the levels in each of the three rooms approached or exceeded the guideline value. It would therefore be expected that the personal exposure of the occupants present would have also approached if not exceeded the WHO guideline value.

As a comparison, at the DoE outdoor monitoring station in Central London (Victoria) in 1993 (during the period when many of the BRE measurements were made), there were no exceedances of the WHO guideline for the outdoor air (12).

Passive and Continuous Detector Measurement of Average Exposure

Palmes tube measurements:

Table 2 shows the average weekly NO_2 concentrations measured by the LMA-3 detectors and the Palmes diffusion tubes. The Palmes tubes readings have been segregated to indicate which batch of tubes was used in each home. The batch date refers to the date the tubes arrived at BRE.

As previously mentioned, initially a single Palmes tube was placed in each room as the tubes had an expected accuracy of better than 10%. For Homes D and E, two tubes were placed at each location to compare results obtained from tubes in the two batches. As can be seen in Table 2, some large variations were observed between these Palmes tube measurements. This was thought to be due to the tubes in batch 1 having exceeded their storage life (initially quoted as six months and later reduced to three months) and therefore some of the results could be questionable.

For Homes F and G single measurements were again taken, but because of some large differences between the readings from the LMA-3 detectors and the Palmes tubes, it was decided that duplicates should be used for homes H, I and J. As Table 2 shows, large discrepancies were observed between these duplicate sets of measurements. Homes K and L used tubes from batch 3. As can be seen, the readings are reasonably consistent.

Linear regression:

Linear regression analysis was performed to compare the measurements from the two types of detectors. In this analysis, the Palmes tube measurements were taken as the independent variable (x-axis) and the LMA-3 results as the dependent variable (y-axis). A summary of the results is shown in Table 3. All duplicate Palmes tube measurements from the same batch were averaged.

The first trial is an analysis over the total data set. As can be seen the correlation is quite poor for this type of comparison. To understand this, further analysis was performed by splitting the data into smaller subsets. The most interesting results were obtained when the data was separated into the different Palmes tube batches. As can be clearly seen, the correlation between the LMA-3 and Palmes tube results is much better for batches 1 and 3 than for 2 (although the correlation for batch 3 is only close to being significant because of the low number of data points).

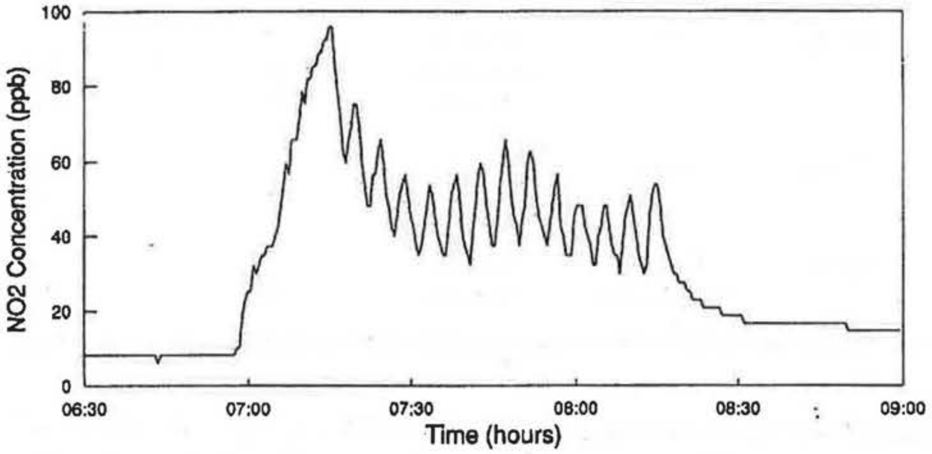

Figure 2. NO_2 Levels from a Central Heating system

Table 2. Passive and continuous measurement of average exposure.

House	Start Date	Cooking Fuel	Location	Average LMA-3 Reading (ppb)	Palmes Tube Reading (ppb)				
					Batch 1 (Dec 92)	Batch 2 (Aug 93)		Batch 3 (Feb 94)	
A	25/5/93	Gas	Kitchen	38	29.6				
			Living Room	16	17.8				
			Bedroom		13.0				
			Out Front		23.7				
			Out Back		19.0				
B	15/6/93	Gas	Kitchen	22	30.0				
			Living Room	14	17.5				
			Bedroom	18	22.5				
			Out Front		8.7				
			Out Back		8.7				
C	22/7/93	Gas	Kitchen	21	26.6				
			Living Room	15	16.6				
			Bedroom	16	16.6				
			Out Front		15.5				
			Out Back		16.6				
D	6/8/93	Gas	Kitchen	18	20.1	28.9			
			Living Room	15	17.6	20.1			
			Bedroom	11	10.1	21.4			
			Out Front		34.0	18.9			
			Out Back		13.8	20.1			
E	23/8/93	Gas Hob	Kitchen	13	7.9	6.7			
		Electric	Living Room	9	6.7	9.0			
		Grill	Bedroom	4	3.3	14.6			
		and	Out Front		18.0	14.6			
		Oven	Out Back		13.5	20.2			
F	21/9/93	Electric	Kitchen	12		24.0			
			Living Room			9.6			
			Bedroom	8		4.8			
			Out Front			26.4			
			Out Back			18.0			
G	5/11/93	Electric	Kitchen	3		3.8			
			Living Room	0		12.8			
			Bedroom	3		6.4			
			Out Front			11.5			
			Out Back			17.9			
H	18/11/93	Gas	Kitchen	32		26.7	35.5		
			Living Room	25		25.2	17.8		
			Bedroom	15		16.3	17.8		
			Out Front			17.8			
			Out Back			37.0			
I	17/1/94	Gas	Kitchen	22		20.6	18.2		
			Living Room	13		10.9	29.1		
			Bedroom	10		12.1	15.7		
			Out Front			53.3			
			Out Back			13.3			
J	7/2/94	Gas	Kitchen	17		14.1	36.0		
			Living Room	10		24.4	42.4		
			Bedroom	2		6.4	23.1		
			Out Front			80.9	15.4		
			Out Back			60.4	19.3		
K	21/2/94	Gas	Kitchen	25				24.4	21.8
			Living Room	11				7.7	9.0
			Bedroom					10.3	9.0
			Out Front					20.5	
			Out Back					18.0	
L	12/5/94	Gas Hob	Kitchen	33				37.2	32.7
		Electric	Living Room					26.7	25.3
		Grill	Bedroom	21				25.3	29.7
		and	Out Front					19.3	22.3
		Oven	Out Back					14.9	35.7

It suggests that there may have been some error in the preparation of the tubes from batch 2. Combining batches 1 and 3 together, gives a fairly good correlation of 0.89. Thus r^2 equals 79%, which means 79% of the variance in the LMA-3 results can be explained by the Palmes tube measurements. The other 21% is presumedly explained by inaccuracies and differences in the measurement techniques. For the final trial, the intercept was forced through zero. Because of the error on the intercept, r reduced very little. It is interesting to see that there is then approximately a ratio of 1:1 between the measurement of NO_2 by the two techniques. Figure 3 displays this data and shows the regression fitted line of best fit. The regression analysis also provided the standard error on the Y estimate. This was 4ppb, i.e. given data from the Palmes tubes, the regression allows the corresponding LMA-3 reading to be estimated with a standard error of 4ppb.

Boliej et al (13) performed a comparison between tubes and a conventional chemiluminescent monitor in 9 homes (one home twice) for a period of between 3 and 12 days in the kitchen, living room and bedroom. For this study, duplicate Palmes tubes were used. The overall coefficient of correlation, r^2, was 0.86. This value is slightly larger than in the present study. This is quite easily explained by their use of duplicate Palmes tubes, which aids greater accuracy of measurement, and the larger number of data points, 30 in total. Using the data provided in the paper, and forcing a zero intercept lowered r^2 slightly to 0.84 and gave a ratio of Palmes tube to monitor concentration of 1.15.

Campbell *et al* (14) performed simultaneous measurements of outdoor air using both diffusion tubes (results used mean of four tubes) and conventional chemiluminescent monitors. A tube to monitor concentration ratio of 1.3 was obtained (no quoted degree of correlation). However, the authors suggest that the diffusion tubes may have read high because of wind effects. This would not affect indoor measurements.

Comparison of Peak to Average Data

It would be useful to have a cheap and practical means of measuring short-term exposure to peak concentrations of NO_2, in a large number of homes. Unfortunately Palmes diffusion tubes cannot be used as they do not have sufficient accuracy. In addition, there are no electro-chemical sensors on the market (which are small, relatively inexpensive and easy to use and maintain) with sufficient accuracy for measurement in the ppb range. Chemiluminescent monitors are too expensive and impractical for this purpose. With this in mind, it was investigated whether there was any correlation between the integrated exposure measurements obtained by the Palmes tubes and peak measurements obtained by the LMA-3 detectors at the same location. For this analysis only tubes from batches 1 and 3 were used.

Linear regression analysis between the Palmes tube readings and the maximum one hour average, gave a correlation coefficient, r, of 0.88 ($p<0.01$). The form of the regression fitted line was as follows:

LMA-3 Maximum 1hr Average = 8.0 x Palmes Tube Reading - 26

Forcing the fit through `zero' reduced the correlation coefficient, r, slightly to 0.87, and the form of the regression fitted equation became:

LMA-3 Maximum 1 hr Average= 6.8 x Palmes Tube Reading

The regression fitted data is shown in Figure 4. It can be seen that at low Palmes tubes readings the data is predominantly below the fitted line whilst for high Palmes tube readings the data is now mainly above the line. Further analysis of the data provided an explanation for this. Many of the

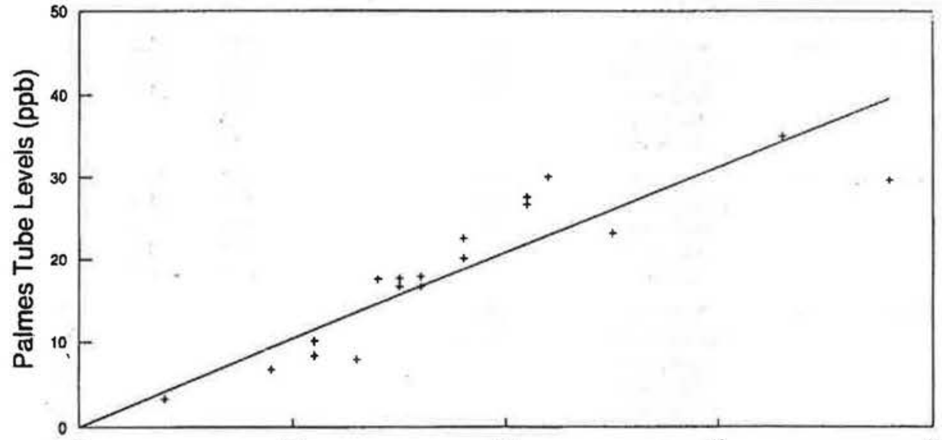

Figure 3. Linear Regression between LMA-3 and Palmes Tube Data Batches 1 & 3

Table 3. Linear Regression Between LMA-3 and Palmes Tube Average Exposure Levels

Tubes	Number of Data Points	Linear Regression		r	p
		Gradient	Intercept		
All	38	0.74	7.0	0.73	< 0.01
Batch 1	14	0.91	2.4	0.86	< 0.01
Batch 2	20	0.64	9.5	0.62	< 0.01
Batch 3	4	1.15	-2.3	0.93	0.065
Batches 1 and 3	18	0.97	1.4	0.89	< 0.01
Batches 1 and 3	18	1.04	0.0	0.89	< 0.01

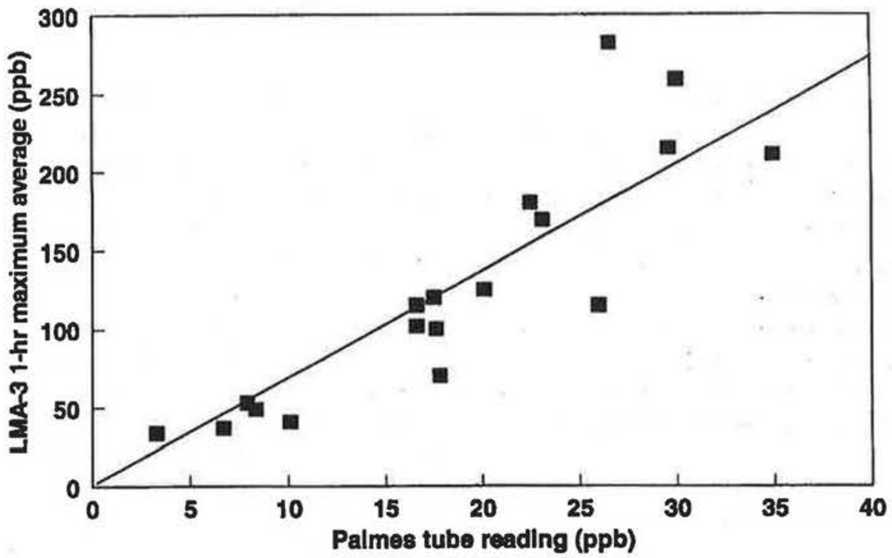

Figure 4. Linear Regression between Peak and Average Data

highest readings correspond to data recorded in the kitchen, where, as is to be expected, the peak to average ratio is higher than that in the living room and bedroom. Similar results have been observed by Lebret *et al* (15).

The WHO 1-hour guideline is 210ppb. Even with the scatter in the data shown in Figure 4, these results suggest that Palmes tube measurements could provide a reasonable prediction of whether the 1-hour guideline has been approached or exceeded during the course of the tube's exposure period and whether additional monitoring is necessary. Further work is required to validate this work, with a larger sample size. However it does suggest that in any large Palmes tube study it may be worthwhile to perform continuous measurements in a number of representative homes which, from the resulting regression analysis, will allow peak concentrations to be estimated for the rest of the homes.

Investigation of NO_2 Removal from a Kitchen with an Extractor-Type Cooker Hood

During the monitoring of Home L, additional information was supplied in the diary of cooker usage to report when the extractor-type cooker hood was used and whether the internal kitchen door was open or closed. Gas cooking took place six times during the week's monitoring and the measured NO_2 levels are shown in Figure 5. Each of the six cooking `periods' shown in Figure 5 is three hours long. On each occasion, gas cooking lasted between 45 minutes and an hour. The peaks correspond only to hob use as the oven and grill are electric. The cooker hood was always set at `high'.

The results showed that during periods 1 and 6, when the cooker hood was not used, prolonged elevated peaks were observed. By comparison during period 3, when the cooker hood was on and the kitchen door was closed, lower levels

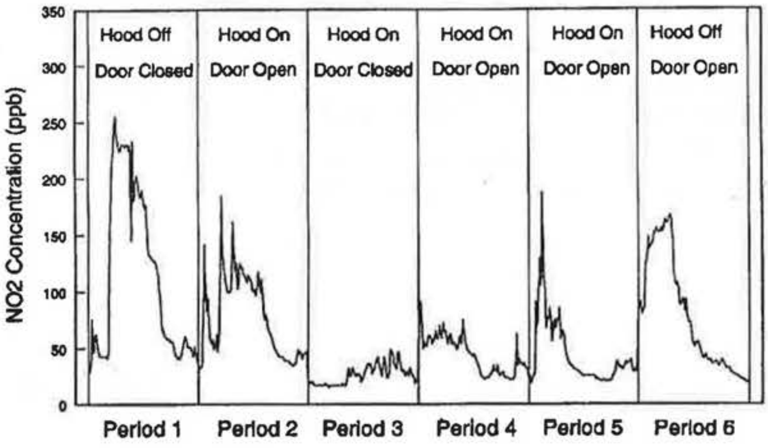

Figure 5. NO_2 Removal by a Cooker Hood

of NO_2 were recorded. In periods 2, 4 and 5 the cooker hood was on and the kitchen door was open. On average, during these othree periods lower levels were seen than with the cooker hood switched off. Transient peaks were observed, probably as a result of the kitchen door being open, resulting in fluctuating ventilation rates and influencing the effectiveness of the cooker hood.

These results do suggest that there could be an advantage of using a cooker hood to maintain lower concentrations of NO_2 during cooking, especially if there are no other competing means of ventilation.

CONCLUSIONS

There are a number of conclusions to be drawn from this work and they can be summarised as follows:

(i) Gas cooking was the principal cause of peak levels of NO_2 in the homes monitored.

(ii) The levels of NO_2 exceeded the WHO one-hour guideline in six of the ten homes using gas for cooking. Levels in homes with electric cooking were well below the guideline value.

(iii) Personal exposure in two of the homes is expected to have approached or exceeded the WHO 1-hour guideline.

(iv) Linear regression analysis on the average exposure levels recorded by the Palmes tubes and the LMA-3 detectors gave a fairly good correlation between the two sets of measurements. The ratio of Palmes tube to LMA-3 concentrations was 1.04. These results are comparable with those of other studies.

(v) Linear regression analysis showed that there was fairly good correlation between the averaged exposure values recorded by the Palmes tubes and the maximum 1-hour averaged levels recorded by the LMA-3 detector within the same room. This suggests that it may be possible to predict whether the 1-hour WHO guideline has been approached or exceeded during the course of a tube's exposure period.

(vi) An investigation of the effect of a cooker hood on NO_2 levels in a kitchen suggests that cooker hoods may make an effective contribution to the removal of NO_2 as well as moisture.

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REFERENCES

1. Raw G. and Coward S., Exposure to nitrogen dioxide in homes in the UK: a pilot study. *Building Research Establishment Occasional Paper*, OP46, (1992).
2. Atkins D.H.F, Healy C. and Tarrant J.B., The use of simple diffusion tubes for the measurement of nitrogen dioxide levels in homes using gas and electricity for cooking. *Harwell*: AERE, Report No. R9184, (1978).
3. Melia R.J.W., Florey C.duV., Morris R.W., Goldstein B.D., Clarke D., John H.H., Craighead I.B. and Mackinlay J.C., Childhood respiratory illness and the home environment: II. Associations between respiratory illness and nitrogen dioxide, temperature and relative humidity. *Int. J. Epidemiol.*, 11, 164-169,(1982)
4. Gardner D.E., Miller F.J., Illing J.W. and Graham J.A., Non-respiratory function of the lungs: host defences against infection. In: *Air Pollution by Nitrogen Oxides*, Schneider T. and Grant L. (eds.), Elsevier Scientific Publishing Company, Amsterdam, 401-415, (1982).

5. Miller F.J., Graham J.A., Raub J.A., Illing J.W., Menache M.G., House D.E. and Gardner D.E., Evaluating the toxicity of urban patterns of oxidant gases. II. Effects in mice from chronic exposure to nitrogen dioxide. *J. Toxicol. Environ. Health* **21**(1-2), 99-112, (1987).
6. Stevenson K.J., Measurements of carbon monoxide and nitrogen dioxide in British homes using unflued heating or cooking appliances. *Tokia J Exp Clin Med,* **10**(4) , 295-301, (1985).
7. Scintrex/Unisearch, LMA-3 Luminox Operators Manual. Concord, Ontario (1985).
8. Palmes E.D., Gunnison A.F., DiMattio J. and Tomczyk C. Personal sampler for NO_2. *Am. Ind. Hyg. Assoc. J.*, **37**, 570-577, (1976).
9. Cadoff B.C., Knox S.F. and Hodgeson J.A., Personal exposure samplers for nitrogen dioxide. National Bureau of Standards, (1979).
10. Apling A.J., Stevenson K.J., Goldstein B.D., Melia R.J.W. and Atkins D.H.F., Air pollution in homes - 2. Validation of diffusion tube measurements. *Warren Spring Laboratory*, Report LR 311 (AP), (1979).
11. World Health Organisation. "Air quality guidelines for Europe". WHO regional publication, European Series No. 23. Copenhagen: World Health Organisation, (1987).
12. Air Monitoring Group. Warren Spring Laboratory, (1994).
13. Boleij J.S.M., Lebret E., Hoek F., Noy D. and Brunekreef B., The use of Palmes diffusion tubes for measuring NO_2 in homes. *Atmos. Environ.* **20** (3), 597-600, (1986).
14. Campbell G.W., Cox J., Downing C.E.H., Stedman J.R. and Stevenson K., A survey of nitrogen dioxide concentrations in the United Kingdom using diffusion tubes: July to December 1991. *Warren Spring Laboratory*, Report LR 893 (AP), (1992).
15. Lebret E., Noy D., Boley J. and Brunekreef B., Real-time concentration measurements of CO and NO_2 in twelve homes. In: *Proc. 4th Int. Conf. on Indoor Air Quality and Climate*, Vol. 1, 435-439, (1987).