SAFETY

HUMAN EXPOSURE TO RADON DECAY PRODUCTS

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SYNOPSIS

After discussing the variation in exposure to radon decay products, the author considers some of the different ways of reducing the harmful effects including increased ventilation.

Introduction

Much concern has been expressed about the radiation exposures resulting from radioactive discharges from the nuclear industry. Yet, on average, only about 0.1% of the average person's exposure is due to this practice. The majority, according to current estimates,[1] somewhat less than 90%, of the average person's exposure is received from natural radiation sources, the remainder being largely due to medical procedures and fallout from weapons tests. The relative magnitude of exposures from natural radiation sources has been used, some might say over-used, by the nuclear industry in an attempt to try to allay public anxiety about radioactive waste discharges and, indeed, they do provide some sense of perspective. But their relative magnitude and, in some cases, extreme variability, justify a deeper consideration of them than merely for the purpose of developing information to support the use of nuclear energy.

It is assumed in radiological protection that any exposure to radiation might cause harm to the exposed person or his descendants. This assumption should apply to exposures from sources of both artificial and natural origin; there is, after all, no intrinsic difference between the radiations emitted by them and their biological effects. Indeed, it is this fact that makes the comparison of exposures from natural and artificial sources valid. Yet, in sharp contrast to the strict controls over exposure to artificial radiation sources, relatively little attention has been given to the control of exposures due to natural radiation sources. There are various reasons for this; an innate feeling that what is natural is without harm or, at least, acceptable; a lack of recognition that exposures may be controllable; and, undoubtedly, a lack of public concern about them.

It is definitely not the purpose of this paper to create public anxiety about natural radiation exposures but rather to point out that there are circumstances where exposures could, and probably should, be reduced. This is particularly so in the case of indoor exposure to radon decay products, which is the subject of this paper.

Sources and Exposures

Apart from radon decay products, sources of exposure to natural radiation include cosmic rays, terrestrial gamma-rays and radioactive material in the body. The National Radiological Protection Board routinely assembles and analyses data on all sources of human exposure to ionising radiation in the UK. A summary of the data from the latest review[1] is given in Table 1; doses from both natural and artificial sources are given. The dose quantity employed is the effective dose equivalent, which provides a common index of risk for uniform and non-uniform irradiation of the body; it is expressed in sieverts, symbol Sv. The unit used in Table 1 is microsievert, μSv, which is a millionth of a sievert.

The values for terrestrial gamma-rays and for radon and thoron decay products are currently under review and may be revised in the near future. Nevertheless, the table reveals that radon decay products are the single most important source of exposure of the UK population.

Origin of radon

Radon is a gaseous radioactive element with many isotopes. Two of consequence occur in nature, radon-220 and radon-222. The first isotope, radon-220, is known as thoron because it occurs in the thorium decay series. The second, radon-222, which is commonly called radon, occurs in the main uranium series. It is considerably more important than thoron in radiological protection because of its longer half-life. Attention here is therefore directed to radon-222 and its common name is used.

Uranium is widely dispersed in the earth's crust at low concentrations. Uranium-238 is the head of the main decay series and its best-known decay product is radium-226, the immediate precursor of radon-222. The amount of radium in earth materials is therefore the direct determinant of the amount of radon generated by them. Since masonry building materials are extracted from the earth, their radium content reflects their origin.

When radon is generated in porous media such as rock and soil or brick and concrete, some enters the pores. Being a gas, radon can move through a medium of this nature, and some is exhaled at the surface. When it is exhaled from the ground out of doors, it is dispersed in the atmosphere, and its concentration in air is low. If radon enters a dwelling, either

Table 1: Annual Average Effective Dose Equivalents to the UK Population.*

Source	Annual Dose (μSv)	Per cent† of total
Natural		
Cosmic	300	14
Terrestrial gamma-rays	400	19
Internal irradiation	370	17
Radon decay products	700	32
Thoron decay products	100	5
Artificial		
Medical procedures	250	11.5
Fallout from weapons	10	0.5
Occupational practices	9	0.4
Radioactive waste disposal	2	0.1
Miscellaneous	11	0.5
Total	2150	100

* Values from the 1984 review.[1] Those for terrestrial gamma-rays and radon and thoron decay products may be revised in the near future.
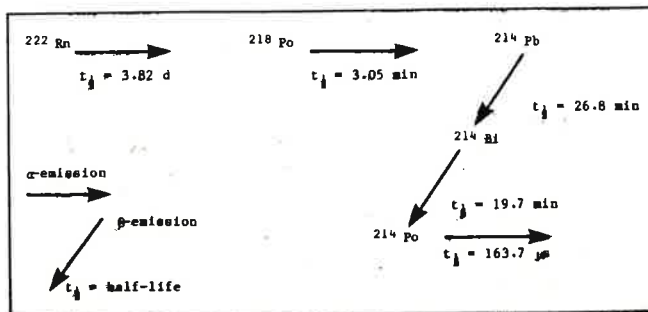† Values rounded for ease of assimilation.

Figure 1: The immediate decay products of radon-222.

Table 2: Radon Levels in UK Dwellings from the National Survey.

Region	Number of Dwellings	Mean Radon Concentration (Bq m⁻³)	
		Living area	Bedroom
UK	2000	28	20
London	123	13	11
Edinburgh	7	16	13
Cardiff	10	35	20
Belfast	3	12	9
South-west England*	50	80	63
Eastern England†	95	26	18
Aberdeen	9	18	10

* Cornwall and Devon
† Norfolk and Suffolk

Table 3: Radon Levels in the Living Room of Dwellings in Selected Areas.

Selected Areas of:	Number of Dwellings	Mean Radon Concentration (Bq m⁻³)
Cornwall	329	390
Devon	150	210
Swansea	7	14
Central uplands*	150	180
Scotland†	170	42

* Derbyshire and Yorkshire
† Dumfries and Galloway, Grampian, and Highland regions

from the ground on which it is built or from the building elements, the concentration increases because dispersion is restricted by the limited ventilation. In underground mines also radon levels are higher for the same reason but attention here is confined to the indoor environment.

Radon decay products

The immediate decay products of radon-222 are radioactive isotopes of solid elements and have short half-lives. The decay chain is shown in Figure 1. On inhalation, the decay products may become lodged in the lung and the effective dose equivalent received is almost two orders of magnitude greater than that from the radon gas alone from which they were produced. It is therefore acceptable to narrow attention to the decay products. Two of the decay products emit beta particles and gamma-rays and two emit alpha particles. The alpha radiation is, however, responsible for most of the radiologically significant dose.

The concentration of radon decay products in indoor air is determined by the radon gas concentration, the ventilation rate and the rate of removal of the decay products to surfaces. Since radon concentrations are higher indoors than outdoors and, on average, only 10% of a person's time is spent outdoors, most of the exposure from radon decay products arises indoors.[2]

National and Regional Surveys

Until recently, there was considerable uncertainty in the UK about the doses to members of the public from radon decay products in dwellings. In the early eighties it began to emerge that there were quite high levels of radon decay products in dwellings in one region of the country and the question was raised whether standards might be necessary to limit exposures. It was recognised at the outset that it would be imprudent to bring in controls without fully understanding existing conditions. Systematic surveys were therefore started,[3] the objectives of which were:

(1): to determine the general distribution of radon concentrations in dwellings, to search for correlations between the measured levels and a number of relevant factors, and to improve the estimates of exposure of the population;

(2): to determine the magnitude of individual exposures and the likely number of dwellings in which exposures might be considered unduly high so that the need for standards could be assessed.

Gamma radiation levels were also to be measured for the sake of completeness.

The first set of objectives is being achieved by a national postal survey of about 2000 dwellings selected systematically from the UK housing stock. Gamma-ray dose rates and average radon concentrations in the living area and main

bedroom are being determined and participants are asked to complete a questionnaire which will provide information on the characteristics of the house and other related matters. Radon concentrations are measured because there is no simple technique available for measuring radon decay product concentrations in a postal survey. Information from the questionnaires and other studies will be used to convert the measurements into radon decay product exposures.

Indoor radon levels

Although the surveys themselves have now virtually finished, a full analysis of the data has yet to be carried out and only provisional results can be given here. A summary of the latest results at present available from the national survey is given in Table 2. To give some feel for the general variations in levels throughout the UK the national survey data have been examined for regional differences and the results are included in the table.

In some cases the sample size is quite small and this limits the accuracy of the estimates of the means. Thus all that can be said about the levels recorded for Edinburgh, Cardiff, and Belfast is that they do not differ greatly from the national average. The sample sizes for London, south-west England and eastern England are more substantial and the differences in levels are statistically significant. Thus the radon concentrations in London are on average about half the national average values, in eastern England about equal to them, and in south-west England about three times higher. These differences are largely a reflection of the differences in local geology. London clay is fairly low in uranium and relatively impermeable to the movement of radon through the ground. The chalks and glacial deposits in eastern England, though relatively low in uranium, are fairly permeable and the granites and neighbouring rocks of south-west England are both relatively high in uranium and fractured.

The results for Aberdeen, although few in number, are interesting. For many years, radiation levels in this city have been considered to be some of the highest in the UK and therefore extensively used for the purpose of comparison with those resulting from the operation of nuclear facilities. Although not given in the Table, the results from the survey show that the gamma-ray dose rates are indeed somewhat above the national values, being, on average, about 40% higher. This is due to the extensive use of granite as a building material. The results in the Table, however, show the radon levels are, if anything, below the averages for the UK. The reasons for this are as yet unclear but several possible causes are cited; low permeability of the granite and sealing of the solum combined with good underfloor ventilation.

The provisional results from the surveys in those areas expected to yield above average levels are given in Table 3. Although referred to by the regions in which they occur, these areas are only parts of those regions and the results should not be considered as representative of those regions as a whole. The results given are those obtained during visits to the houses. The full results of follow-up measurements over long time
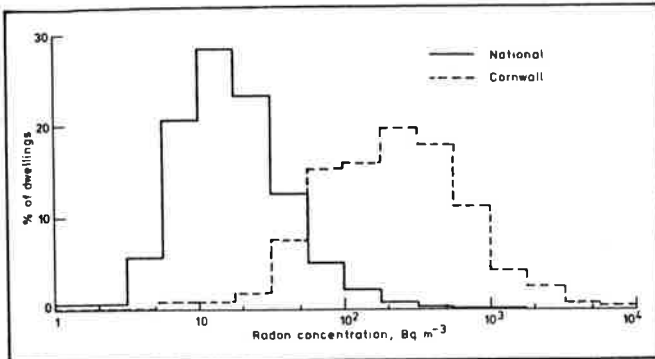
Figure 2: Frequency distribution of radon concentration in air in the living areas of some UK dwellings.

Table 4: Annual Effective Dose Equivalents in UK Dwellings from Radon Decay Products.*

	Annual Effective Dose Equivalent (μSv)	
	UK	Cornwall
Arithmetic Mean	430	6 600
Geometric Mean	250	3 400
95% fractile	1200	22 000
99% fractile	2400	50 000

* Provisional data.

periods are not yet available but will provide better estimates of exposures. At this stage, however, it can be said that the average concentration in the houses in Cornwall that were studied is more than an order of magnitude above the national average level. The levels in the houses in Devon and the Central Uplands are also significantly higher.

The spread of values in these areas is of greater interest than the actual means. The frequency distribution of the data for the areas of Cornwall studied is shown in histogram form in Figure 2 along with the UK data from the national survey for comparison purposes. The results show that the levels in some dwellings in Cornwall are more than two orders of magnitude above the national average.

Radon exposures and risks

A number of assumptions are required to convert radon concentrations into annual effective dose equivalents from radon decay products but these will not be discussed here. Suffice it to say that an average indoor radon concentration of 1 Bq m^{-3} is currently considered to result in an annual effective dose equivalent of about 20 μSv.[5] This conversion factor was used to derive the values given in Table 4. Confirmation of the lower mean exposure to radon decay products than the value given in Table 1 will need to await the final set of results from the national study. Nevertheless, at this stage, the following fairly firm conclusions can be drawn. For the inhabitants of the vast majority of dwellings in the UK, the annual effective dose equivalent from radon decay products does not exceed 2400 μSv. However, in the areas of Cornwall studied, the dose in almost 5% of the dwellings exceeds 25 000 μSv and in 1% of the dwellings 50 000 μSv.

There is no direct epidemiological evidence that exposure to natural radiation in dwellings causes harm to human beings. The risk factors used to calculate the risks from these levels of exposure have been obtained from epidemological studies of various groups such as the survivors of the Hiroshima and Nagasaki atomic bombings and early uranium miners exposed to radon decay products. The basic assumption is made that the risk factors (health consequence per unit radiation dose) are constant irrespective of dose or dose rate. The overall risk factor for fatal cancers and serious hereditary defects currently recommended by the International Commission on Radio-

Table 5: Lifetime Risks of Premature Death from Various Causes including Lung Cancer from Radon Decay Products.

Cause*	Lifetime risk
All malignant neoplasms	2 x 10^{-1} (1 in 5)
Lung cancer from 100 000 μSv a year	7 x 10^{-2} (1 in 14)
Lung cancer from 10 cigarettes a day	6 x 10^{-2} (1 in 17)
Lung cancer from 50 000 μSv a year	4 x 10^{-2} (1 in 25)
Lung cancer from 25 000 μSv a year	2 x 10^{-2} (1 in 50)
Accidents in the home	9 x 10^{-3} (1 in 110)
Accidents on the road	8 x 10^{-3} (1 in 125)
Lung cancer from 5000 μSv a year	4 x 10^{-3} (1 in 250)
Accidents to men at work	2 x 10^{-3} (1 in 500)
Cancer from average public exposure to nuclear waste (2 μSv a year)	1 x 10^{-6} (1 in 1 000 000)

* The radiation units here refer to the effective dose equivalent.

logical Protection[6] is 1.65 x 10^{-2} Sv^{-1} (or 1 in 60 per Sv). This risk factor can be used to calculate the risk of lung cancer from exposure to radon decay products.

This risk factor applied to a simple model of latency and tumour expression can be used to compute the lifetime risk of lung cancer from radon decay products.[7] The lifetime risks of premature death for a range of annual doses from radon decay products are given in Table 5: risks of death from other causes are included for comparison. It can be seen, for example, that the lifetime risk of lung cancer from an annual effective dose equivalent of 5000 μSv is 4 x 10^{-3} (or 1 in 250), which is about 2% of the overall risk of fatal malignancy facing the average individual or about 0.4% of the risk of death from all causes.

Remedies and Prevention

The concentrations of radon and its decay products can be reduced by increasing the ventilation. A simple measure such as the removal of weather-stripping may be sufficient where high radon levels are caused by unusually low ventilation rates. In other cases, electric fans might be installed to increase ventilation, at least as a temporary measure. There is however a penalty to be paid in energy consumption, and it is doubtful whether occupants would be prepared to use such methods on a long-term basis. Ventilation can also be increased in a controllable fashion by mechanical ventilation with heat recovery. Such devices are relatively expensive to install, and it is unlikely that they would find wide-scale application in the UK. In addition, they are likely to be of value only when high radon levels are primarily caused by low ventilation.

Electrostatic precipitators or other devices for aerosol filtration could also be used to reduce the concentration of radon decay products. The capital and running costs would be substantially lower than advanced mechanical ventilation systems. The biggest problem with this technique, as with extractor fans, is that they are active rather than passive and can be regarded only as temporary measures.

Experience, particularly in Sweden, demonstrates that the caulking of apertures in the floor slab can be highly effective in reducing radon ingress and can be carried out at low cost.[8] The use of a barrier to the convective flow of soil gas through cracks in the slab has also been successful in a UK dwelling.[9] These passive methods have the advantage that little or no maintenance is required and the occupant does not need to be continuously aware of the radon problem.

Work in the UK[9] and Sweden[10] also demonstrates that mechanical devices to remove radon from the underfloor space are very effective and can be installed at modest cost. These devices, usually fans, suffer again from the disadvantage that occasional maintenance is required and the occupant must be prepared to operate them.

Some of these remedial techniques could also be adopted as preventive measures during the construction of new dwellings
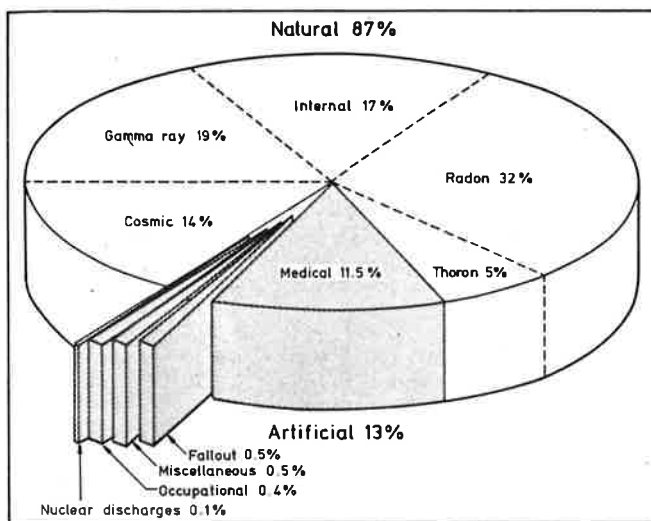
Figure 3: The composition of the total radiation exposure of the UK population.

on sites that would otherwise cause high exposure indoors. It would, in fact, be considerably easier to use appropriate barriers to prevent or reduce substantially the radon that might enter new dwellings from the ground than to remedy existing dwellings on the same site. Modest changes to present design and construction practices are likely to be sufficient to minimise or virtually eliminate discontinuities in the structural membrane separating the dwelling space from the ground underneath.

Protection Standards

The mean dose to the UK population of 430 μSv in a year (Table 4) implies a hypothetical annual rate of death of about 400 per year if the usual assumptions are made that all doses have an associated risk. In the short term, there is little that can be done to reduce this. In the longer term, consideration might be given to the establishing of design standards for future dwellings that would limit radon ingress from the ground.

At the present time, the main objective of any control policy must be to prevent individuals from receiving too high doses. Members of staff of the National Radiological Protection Board put forward suggestions regarding the limitation of dose.[11] A distinction was made between existing dwellings, for which action would be remedial, and new dwellings, for which prevention could be incorporated at the design and construction stages. The suggestions were that there should be an action level of 25 000 μSv a year for existing dwellings and a design level of 10 000 μSv a year for new dwellings.

There is no absolute justification for these levels. The action level of 25 000 μSv a year for instance was considered appropriate since continuous exposure at this level implies an increase approaching 10% in the lifetime risk of contracting fatal cancer and an average risk of death comparable to that from all common accidents combined. Some might suggest lower action levels but it must be remembered that the lower the level, the larger will be the size of the problem and the overall cost to the country. The estimated maximum number of dwellings that might be effected by an action level of 25 000 μSv a year is about 2000; at 5000 μSv a year, the number would be about 50 times higher. Furthermore, the law of diminishing returns applies; as the action level is lowered, so the cost per unit dose saved will increase.

The suggestions by members of NRPB staff were considered by the Royal Commission on Environmental Pollution.[12] The recommendations of the RCEP given in its tenth report, which was published last year, are reproduced in full here.

(1): Priority should be given to devising effective remedial measures for those existing houses where radon concentrations are such as to cause anxiety, and to devising preventive measures for new houses.

(2): Remedial action should be undertaken on existing dwellings, initially on those where the annual effective dose

equivalent exceeds 25 mSv (25 000 μSv).

(3): A design limit should be applied to new dwellings. On the basis of information available to us we favour an annual effective dose equivalent limit of 5 mSv (5000 μSv), but a decision on the value should await the assessment of recommendations in the forthcoming report on controllable sources of radiation by the International Commission on Radiological Protection.

Since the RCEP report, the ICRP recommendations have been published[13] and, although there are some differences, largely of emphasis, they are broadly in line with the NRPB staff suggestions.

That is essentially where the position on the development of standards rests at the present time. Before the NRPB, as part of its statutory functions, can offer formal advice to Government, the full and considered results of the surveys of radon in dwellings and the studies of remedial and preventive techniques are required. It is expected that this information will become available during 1986.

Implementation

There is much less concern and hence less interest in radiation exposures of natural origin than of artificial origin and unless some public awareness of the significance of indoor exposure is generated, it is possible that any action to deal with the problem will be misunderstood. It is necessary therefore to develop a systematic approach to public education about radon in dwellings. The aims of a programme of information should be to indicate that there is some risk from radon decay products, that measures are feasible to reduce the risk, that the problem is not acute even for high levels, but that action is required without undue delay. To this end, the NRPB has deliberately followed a policy of providing full information on the national and regional survey work and on the remedial studies. This information has been widely reported in the media, and further material will be released as the data are being analysed. But dissemination of information on potential health hazards is not without its own risks as it can lead to undue public anxiety or the accusation that one is merely playing upon human fears.

The procedure for implementing standards has yet to be determined in the UK, and no decision has been taken whether statutory regulations or administrative procedures should be used: indeed a combination of both may be most appropriate. This is a matter for the appropriate Government department rather than NRPB, but it is well to recognise that there are considerable difficulties in effecting controls over the environment in existing dwellings.

For new dwellings, the situation is, in principle, more straightforward. Building regulations apply to the construction of dwellings, and it would seem that, if they were deemed appropriate, they could be used to avoid a hitherto unrecognised hazard. Since radon levels in dwellings are affected by the type of floor or the use of radon barriers in the floor, and since these are matters that could readily be inspected, implementation should not pose serious problems.

Whatever procedure is used for implementing standards, it will be necessary to establish methods for discovering dwellings with high radon levels. Systematic measurements will need to be mounted in all likely dwellings in areas expected to yield high results using simple inexpensive monitoring techniques. The detection of suspect building sites would need to be based on an assessment of geolgical information, such as the type of ground and its permeability, and a study of the results of any previous surveys of radon levels in dwellings in the vicinity.

Conclusion

Some risks of everyday life, such as those from driving and flying, are obvious; others, such as those from smoking, can only be inferred from statistical information. The absence of

Events
1986

22-26 September: **Diffusive Sampling.** Luxembourg.

This symposium is being organised by the Commission of the European Communities and the United Kingdom Health and Safety Executive, with the participation of other international organizations.

There is an increasing requirement to monitor worker exposure and the working environment for airborne contaminants within an overall strategy to protect worker's health. This has led to the development of new, effective and less costly sampling systems based on the principle of diffusion.

The objectives of the symposium will be: to review the state of the art of diffusive sampler techniques; to stimulate the exchange of technical information; to assess the suitability and range of applications for workplace monitoring; to promote the further development of this technique and also its wider use.

This symposium is aimed at occupational health physicians and hygienists, analysts, employers, trade-union representatives, regulatory authorities, workmen's compensation boards, research institutes, manufacturers of equipment and all potential users of diffusive samplers.

There will also be an exhibition of sampling and analytical equipment. The official languages will be English and French.

Details from: MORSI, Centre of Information – Documentation, 70 rue de Luxembourg, L-7330 Heisdorf G.-D. Luxembourg.

23-26 September: **Heating and Processing.** Cambridge, UK.

The conference is being organised in association with the International Union for Electroheat, the International Microwave Power Institute and the Microwave Association. It will be concerned with radio frequency and microwave techniques for the heating and processing of non-metallic materials in the frequency range 1 — 3000 MHz. Such techniques can be used in a wide range of industries for heating, drying, curing, cooking, and joining.

The technical sessions, discussions and associated activities will be of interest to equipment designers and manufacturers, process engineers, academics and users interested in heating processes.

Offers of papers are invited. *Details from:* BNCE, 30 Millbank, London, United Kingdom SW1P 4RD.

Large Scale Applications of Heat Pumps
25-27 March 1987
Call for Papers

BHRA, the Fluid Engineering Centre, is to hold its third international conference on the Large Scale Applications of Heat Pumps, in Oxford, England, from 25 to 27 March 1987. Offers of papers are invited.

The conference is expected to cover: the applications of heat pumps; systems studies; unit processes; thermodynamics; district heating and cooling; working fluids; novel types of heat pumps; and the economics involved. Authors are invited to submit titles and synopses of papers to reach the organisers no later than 1 April, 1986.

For further details contact: The Conference Organiser (LSAHP), BHRA, The Fluid Engineering Centre, Cranfield, Bedford, United Kingdom MK43 0AJ. *Telephone:* National (0234) 750422; International + 44 234 750422; *Telex:* 855059.

Courses
1986

A number of short courses on: Lighting; Energy Management; and Building Services are to be held at the Polytechnic of the South Bank, London, England.

Details from: Short Courses Co-ordinator, IoEE, Polytechnic of the South Bank, Borough Road, London, United Kingdom SE1 0AA. *Telephone:* National 01-928 8989 Extension 2112; International +44 1 928 8989 Extension 2112.

HUMAN EXPOSURE TO RADON DECAY

direct epidemiological evidence linking indoor exposure to lung cancer might lead some observers to suggest that standards are unnecessary, and indeed the very thought that natural irradiation indoors may be harmful to health is unpalatable to some persons. There is no doubt, however, that adequate information already exists to establish the potential harm of radon decay products in dwellings, and that levels in some houses are sufficiently high to justify the setting of safety standards.

Work in the UK and elsewhere has shown that methods for reducing high radon levels in existing dwellings and of preventing high levels in future dwellings are feasible and can be implemented without undue cost. There is therefore no impediment to the production of standards on this count. Members of staff of the NRPB have already made suggestions about dose levels in dwellings, the aim being to limit the risks to occupants, and these have been broadly endorsed by the RCEP. These suggestions are also broadly in line with those made more recently by the ICRP.

The approach to formal standards in the UK is being undertaken deliberately and systematically. The results of the national and regional surveys of indoor radon levels have still to be fully analysed and assimilated, and some further work needs to be done on remedial and preventive measures. An efficient means of detecting building sites, which might subsequently cause high indoor levels if precautions were not taken during construction, is still to be devised. These are the radiological considerations on which the NRPB will offer formal advice to government in due course.

On the administrative side, procedures need to be developed for the efficient implementation of standards. Decisions have to be made on the degree of regulation required, and the lines of responsibility will need to be clear. There is also a need to increase public awareness of the matter in a way that does not cause distress or confusion but will facilitate the implementation of the standards.

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TECHNICAL PAPERS

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Indoor Air Pollution Due to Emissions from Unvented Gas-Fired Space Heaters

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Operation of an unvented combustion appliance indoors can elevate pollutant concentrations. Under laboratory conditions, oxygen consumption rates and pollutant emission rates of CO, CO_2, NO, NO_2, HCHO and submicron suspended particles emitted from eight unvented gas-fired space heaters operated with well adjusted air shutters at partial and full fuel consumption rates were determined in a 27-m^3 chamber. Emission rates were also determined for some heaters operating under poorly tuned conditions. Four of the eight heaters were subsequently tested in a 240-m^3 research house with 0.36–1.14 air changes per hour. Based on measurements near steady state, steady state pollutant and oxygen levels were projected: 1930–11,100 ppm for CO_2, 1.0–26 ppm for CO (under well-tuned conditions), 0.40–1.46 ppm for NO_2, and 19.1–20.7% for O_2. Concentrations of CO_2, CO, and NO_2 sometimes exceeded outdoor or occupational guidelines. Analysis showed that CO, NO, and NO_2 emission rates can vary with time and that, while short-term emission rates derived from laboratory tests were consistent with initial emission rates observed in the field, they did not always correspond to steady state emission rates.

It has been demonstrated that operating unvented combustion appliances indoors increases the indoor concentrations of several pollutants.[1-5] One such appliance of current interest is the unvented natural gas-fired space heater (UVGSH). This study examines the pollutant emissions and concentrations from eight UVGSHs under laboratory conditions and from four heaters under controlled-field conditions. The eight UVGSHs tested were purchased in 1981 from three U.S. manufacturers and were not equipped with oxygen depletion sensors. The heaters have a blue flame when well tuned and each heater has removable ceramic inserts, serving as radiant elements, positioned at least 2 cm above the burner.

The goals of this study were to quantify pollutant emission rates in the laboratory, to determine whether these emission rates were applicable to field situations, and to determine representative indoor pollutant levels from use of an UVGSH in a residence. (Indoor pollutant levels from the use of UVGSHs depend on many factors—the appliance's pollutant emission rate, the appliance usage pattern, and building-specific parameters such as volume and air exchange rate.) All heaters were tested for their consumption of oxygen (O_2) and for their emissions of carbon monoxide (CO), carbon dioxide (CO_2), nitric oxide (NO), nitrogen dioxide (NO_2), formaldehyde (HCHO), and submicron suspended particles. This report summarizes more extensive reports of our laboratory and field results.[6,7]

Experimental

All laboratory measurements were conducted in an environmental chamber and all field measurements were conducted in an unoccupied residence.

Most of the instrumentation used for both laboratory and field experiments is contained in our Mobile Atmospheric Research Laboratory (MARL). The eight heaters tested in the laboratory phase of this project had manufacturer's fuel input ratings that ranged from 12,600 kJ/h (12,000 Btu/h) to 42,200 kJ/h (40,000 Btu/h). The fuel input ratings of the four heaters used in the field phase ranged from 12,600 kJ/h (12,000 Btu/h) to 31,600 kJ/h (30,000 Btu/h).

The environmental chamber is a 27-m^3 structure housed within a larger building that buffers outdoor environmental fluctuations. To promote air mixing, six 10-cm low-flow fans are located inside the chamber in a manner that avoids air drafts near the heater.

The field research house is an unoccupied, partially furnished, one story residence with an air volume of 240 m^3. The air sampling sites were located 1.5 m above the floor. The fireplace and all furnace ducts were sealed so that the interior air volume would be well defined. Some doors and windows were slightly opened (from 0.5 cm to 3.2 cm, increasing the leakage area in the building envelope by approximately 400–1400 cm^2) to vary air exchange rates. Total air exchange rates achieved in this manner ranged from 0.36 to 1.14 air changes per hour (ach). No mixing fans were used in the house.

Instrumentation

The MARL is designed for remote multipoint sampling of CO, CO_2, NO, NO_2, O_2, temperature, dew point, and outdoor wind speeds.[6-8] The MARL

sampling system continuously draws air samples through Teflon tubing from four locations and uses a multipoint timing system to automatically switch between sites at pre-set intervals. Teflon filters are placed at the inlets of the sampling lines for protection against particulate matter.

A refrigerated bubbler sampler was used to collect HCHO for subsequent laboratory analysis.[9] Suspended particulate concentrations, for particles between 0.0056 and 0.56 μm in diameter, were measured using an electrical mobility analyzer.[10] Generally, particles were measured every 10 minutes.

Modeling

A single-equation, mass-balance model was used to calculate pollutant emission rates of the UVGSHs from laboratory data using a technique previously published.[8] This model has been used successfully to predict indoor air pollution levels as well as to determine indoor air quality parameters that can affect such levels.[5–8,11,12]

The mathematical expression for a change in the average indoor gaseous pollutant concentration of a whole house is:

$$dC = PaC_o \, dt + \frac{S}{V} \, dt$$
$$- (a + k)C \, dt \quad (1)$$

where: C = indoor pollutant concentration (ppm);
C_o = outdoor pollutant concentration (ppm);
P = fraction of the outdoor pollutant level that penetrates the building shell (unitless);
a = air exchange rate (h^{-1});
S = indoor pollutant source strength (cm^3/h);
V = volume (m^3);
k = net rate of removal processes other than air exchange (h^{-1}).

For particles, C and C_o are usually ex-

pressed in units of $\mu g/m^3$ and S in units of $\mu g/h$. Assuming C_o, P, a, S, and k are constant over the time period of interest, Eq. 1 can be solved for $C(t)$ to give:

$$C(t) = \frac{PaC_o + S/V}{a + k} [1 - e^{-(a+k)t}]$$
$$+ C(0) \, e^{-(a+k)t} \quad (2)$$

Eq. 2 describes the spatial average concentration of a pollutant in an enclosed space of a given volume.

Solving Eq. 2 for S, dividing it by the fuel consumption rate, R (kJ/h), and letting T equal the duration of appliance operation, we can obtain the emission rate, E ($\mu g/kJ$ for particles and cm^3/kJ for gases):

$$E = \frac{S}{R}$$
$$= \frac{V}{R} (a + k) \frac{[C(T) - C(0)e^{-(a+k)T}]}{[1 - e^{-(a+k)T}]}$$
$$- \frac{VPaC_o}{R} \quad (3)$$

For gases, E, in cm^3/kJ, has been converted to $\mu g/kJ$ by using the ideal gas law and the time-weighted average temperature and pressure in the space of concern. Note that Eq. 3 relies on the final average indoor pollutant concentration, $C(T)$. For laboratory tests, the use of mixing fans increases the accuracy and precision of the $C(T)$ measurement.

Laboratory Test Protocol

Prior to tests, all heaters were tuned by adjusting the air shutter for a minimum CO concentration measured just above the heater and by visually observing the flame characteristics. For the partial input tests, fuel consumption rates were set by adjusting the heater's gas control valve to obtain a flame approximately one half its full input height.

After the mixing fans were turned on, indoor and outdoor background measurements were made. Then the heater was ignited and operated until approximately 5500 kJ (5.0 ft^3) of natural

gas was consumed. After the heater was shut off, the decay of pollutant levels in the chamber was monitored for one hour. Data from this decay period were used to calculate the air exchange rate, the reactivity rate, and the peak concentration for each pollutant.[8] A one-hour measurement of average HCHO concentration was made simultaneously inside and outside the chamber during the decay portion of the test. Following the decay period, pollutant levels outside the chamber were measured for 15 minutes.

Field Test Protocol

The measurement protocol for field testing was similar to that of the laboratory tests. The main differences were that the heaters were operated from two to eight hours and three indoor locations and one outdoor location were monitored on a rotating basis.

The UVGSHs were operated in either the living room or the bedroom. CO_2, CO, NO_2, NO, and O_2 concentrations from outdoors, kitchen, living room, and bedroom were recorded every minute on a rotating basis with six minutes at each site. Data generated during this cycle were treated by discarding the first three data points and averaging the last three data points, thus obtaining one "observation" every six minutes, i.e., twenty-four minutes elapsed between observations at a single site. For some data analyses the average pollutant concentration of the whole house was required and the gas-phase pollutant data from the three indoor locations were averaged.

For each test, the average air exchange rate of the house was determined by taking a time-weighted average of the air exchange rate computed from the rise of the CO_2 concentration and the rate determined from the decay of CO_2. The difference between the "decay" and "rise" air exchange rates averaged 0.04 ± 0.10 h^{-1}, not significantly different from zero. The average relative standard deviation for the rise and decay air exchange rates was 10%.

Table I. Pollutant emission rates from eight well-tuned, unvented gas-fired space heaters operated at full input in an environmental chamber with low ventilation.

Heater[a]	Fuel consumption rate (kJ/h)	No. of tests	CO ($\mu g/kJ$)	NO ($\mu g/kJ$)	NO$_2$ ($\mu g/kJ$)	N (of NO$_x$) ($\mu g/kJ$)	CO$_2$ ($\mu g/kJ$)	O$_2$ ($\mu g/kJ$)	HCHO ($\mu g/kJ$)	Particles[c] ($\mu g/kJ$)
12A	10,100	3	114	9.6	20	11	50,000	−67,400	4.2	3.2
20A	22,800	3	29	23	13	14	50,100	−71,700	0.61	0.39
30A[c]	35,400	1	25	22	11	14	49,900	−72,900	0.59	0.06
16B	17,900	1	165	14	18	12	51,500	−71,900	0.55	0.49
40B	44,700	2	63	17	20	14	51,000	−68,900	0.96	0.09
20C	22,800	2	14	16	11	11	50,100	−73,700	0.91	0.79
30C	31,900	1	11	19	9.6	12	52,600	−73,700	0.43	0.64
40C	43,000	2	13	19	10	12	54,600	−71,000	0.61	0.24

[a] Identification code: 40B = 40,000 Btu/h (42,200 kJ/h) heater from manufacturer B. Heat content of natural gas was 39.1 kJ/L.
[b] Mass of particles from 0.0056 to 0.56 μm in diameter analyzed by an electrical mobility detector, assuming a particle density of 2.0 g/cm^3.
[c] Heater subjected to extensive tuning and considered optimally tuned.

Results and Discussion

Laboratory Results

To assess the reproducibility of laboratory emission rate tests, five experiments were repeated in duplicate and two experiments were repeated three times. The seven experiments included tests of heaters under well-tuned and maltuned conditions under low and high ventilation rates and one experiment with the heater under partial fuel input. The means of the relative standard deviations (with the ranges in parentheses) for the pollutant emission rates and O_2 consumption rates in the seven sets of replicate experiments were: 3.0% (0.3–5.0%) for CO_2; 2.6% (1.0–5.3%) for O_2; 35% (5.3–80%) for CO; 14% (2.2–35%) for NO; 15% (6.2–27%) for NO_2; 7.7% (2.0–16%) for N (of NO_x); 25% (17%–39%) for HCHO; and 53% (6.6–83%) for submicron particles. (Only three data sets were used to determine the reproducibility of submicron particulate measurements.)

Since the variations in the CO_2 and O_2 replicate measurements were low, our technique for determining emission rates appears to be precise. The observed variations of the other pollutants appear real since the variations are greater than the precisions of their instrumentation and greater than the relative standard deviations of the CO_2 and O_2 emission rates.

Full input tests on well-tuned heaters. Table I presents the test results for well-tuned heaters operated at full input. (A heater code of 30C represents a 30,000 Btu/h heater from manufacturer C.) "Well-tuned," except when otherwise indicated, denotes the intention of the test rather than an assessment of the results. That is, we attempted to optimize tuning (i.e., minimize CO emissions) by adjusting the air shutter using a hand-held CO monitor. However, after a test was completed, it was sometimes evident that the heater had not been optimally tuned. Except in the case of the 30A heater, no readjustments were made after the initial tuning, based on the assumption that the typical consumer would not have readjusted the heater. The CO_2 emission rates and the O_2 consumption rates were relatively constant for all UVGSHs. The average CO_2 and O_2 emission rates of the individual tests shown in Table I are 51,100 $\mu g/kJ$ and 70,900 $\mu g/kJ$, respectively. The relative standard deviation of the measurements for both CO_2 and O_2 was 3%. Based on the composition of the natural gas used (Pacific Gas and Electric, San Francisco) we calculated a theoretical CO_2 emission rate of 51,000 $\mu g/kJ$ and a theoretical O_2 consumption rate of 73,200 $\mu g/kJ$; both values were consistent with those measured.

The average NO emission rate for the eight heaters was 17.3 ± 4.2 $\mu g/kJ$ and the NO_2 rate was 14.1 ± 4.5 $\mu g/kJ$. The average emission rate for nitrogen of the nitrogen oxides (N of NO_x, where NO_x = NO + NO_2) was 12.6 ± 1.5 $\mu g/kJ$. Although NO_x emission rates were fairly consistent for all eight well-tuned heaters, individual emission rates for NO and NO_2 varied between heaters.

As noted, the CO emission rates were much more variable than the emission rates of other pollutants. Five heaters had CO emission rates of less than 30 $\mu g/kJ$ and the other three heaters had much higher rates, up to 165 $\mu g/kJ$. Another research team investigating natural-gas combustion appliances observed that the CO emission rates across appliances appear to be lognormally distributed.[13] Our results are consistent with that observation. The geometric mean of the CO emission rates is 34 $\mu g/kJ$, and the geometric standard deviation (GSD) is 2.80.

HCHO emission rates may also follow a lognormal distribution because, like CO, HCHO is a product of incomplete combustion. The geometric mean emission rate was 0.81 $\mu g/kJ$ (GSD = 2.04). Heater 12A had the highest HCHO emission rate, averaging 4.2 $\mu g/kJ$, based upon three tests. This heater also had the highest particulate emission rate, averaging 3.2 $\mu g/kJ$, for particles between 0.0056 to 0.56 μm in diameter. The mass median diameter of the particulate emissions for selected experiments ranged from 0.12 μm to 0.19 μm (mean GSD = 2.1). The particulate emission rates from all UVGSHs followed a lognormal distribution with a geometric mean of 0.38 $\mu g/kJ$ (GSD = 3.5).

Partial-input tests on well-tuned heaters. Tests were conducted on UVGSHs operated at partial fuel input. Although all three manufacturers informed us that these heaters were not designed to be used at partial input, we found that all heaters tested operated at partial input without difficulty or apparent deterioration in performance. The range of fuel input adjustment, while not large, allows the user to obtain a more constant indoor temperature. While deemed by the manufacturers to be a misuse of the product, operating at partial input may be common among users.

On average, the emission rate for each pollutant measured, except CO_2, was lower during partial-input operation than during full-input operation. The NO, NO_2, and N (of NO_x) average emission rates were 13.0 ± 2.0 $\mu g/kJ$, 10.2 ± 2.2 $\mu g/kJ$ and 9.2 ± 1.1 $\mu g/kJ$, respectively. The geometric means for CO and HCHO emission rates were 17.2 $\mu g/kJ$ (GSD = 2.1) and 0.52 $\mu g/kJ$ (GSD = 3.3), respectively. The geometric
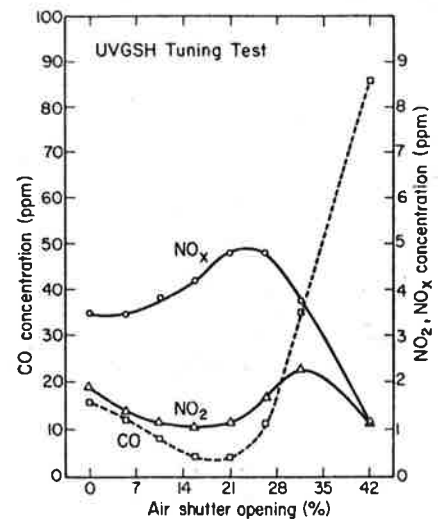

Figure 1. Peak CO, NO_2 and NO_x concentrations versus the primary air shutter opening for a 31,700 kJ/h unvented gas-fired space heater (heater code: 30A) after combusting 142 L (5 ft³) of natural gas in a 27-m³ environmental chamber. The chamber air was well-mixed and the ventilation rate was approximately 0.4 ach.

mean emission rate for submicron particles was determined to be less than or equal to 0.05 $\mu g/kJ$.

Tuning tests. The variability of CO emissions prompted further testing of the sensitivity of emission rates to adjustments of the air shutter. We measured peak CO, NO, and NO_2 concentrations from heater 30A after combusting 142 L (5 ft³) of natural gas under constant ventilation conditions (0.4 ach) with adjustment of the air shutter as the only variable (see Figure 1). Throughout the range of shutter settings depicted, 0 to 42% of fully open, the visual flame characteristics were good. With the air shutter opening increased from 21% to 32% of fully open (only a 10° rotation of the shutter) the CO concentration increased by a factor of 9.

Although NO_x emissions were not quite as sensitive to tuning as the CO emissions, the NO_2 to NO_x ratio was very sensitive, varying from approximately 0.2 at a 21% shutter opening to almost 1.0 at a 42% shutter opening. The NO_2 emissions varied only by a factor of 2 over the shutter opening range of 0% to 42% open. As expected, the minimum CO emission occurs near the same shutter setting as the maximum NO_x production; i.e., the flame is hottest when combustion is most complete. However, finding an NO_2 emission minimum near the CO emission minimum was unexpected.

Additional tests were conducted on the 30A heater with the air shutter completely closed and fully open. With the air shutter closed, the CO emission rate was 159 $\mu g/kJ$, NO was 15.1 $\mu g/kJ$, NO_2 was 13.7 $\mu g/kJ$, HCHO was 1.1 $\mu g/kJ$, and submicron particulate

Table II. Average pollutant emission rates from unvented gas-fired space heaters operated at full input in an unoccupied field research house.

Heaters[a]	Fuel consumption rate (kJ/h)	Number of tests	CO (μg/kJ)	NO (μg/kJ)	NO (μg/kJ)	N (of NO) (μg/kJ)	State of tune
12A	9,510	1	32	24	8.6	14	well tuned
30A	31,400	5	11 ± 6	29 ± 4	7.5 ± 1.9	16 ± 2	well tuned
30A	31,500	3	37 ± 20	22 ± 1	9.0 ± 1.7	13 ± 1	excess fuel
30A	31,900	1	620	<0.1	20	6.1	excess air
16B	16,800	7	190 ± 30	20 ± 3	18 ± 2	15 ± 2	well tuned
30C	31,200	1	12	29	7.4	16	well tuned

[a] Identification code: 30C = 30,000 Btu/h (31,600 kJ/h) heater from manufacturer C. Heat contents of natural gas was 39.1 kJ/L.

emissions were less than 0.04 μg/kJ. With the air shutter fully open, the CO emission rate was 517 μg/kJ, NO was 0.04 μg/kJ, NO_2 was 11.5 μg/kJ, HCHO was 20.3 μg/kJ and submicron particulate emissions were below 0.04 μg/kJ. The very high CO and HCHO emissions with the shutter fully open again demonstrate the importance of tuning on the pollutant emission rate of this type of combustion appliance.

The 40C heater was also tested for pollutant emission rates with its shutter closed and fully open. The results show that this heater was relatively insensitive to tuning compared to the 30A heater. With its air shutter closed, the CO emission rate was 35 μg/kJ, NO was 13.7 μg/kJ, NO_2 was 11.2 μg/kJ, HCHO was 0.22 μg/kJ, and the submicron particulate emission rate was 0.07 μg/kJ. With the air shutter fully open, the CO emission rate was 8 μg/kJ, NO was 19.9 μg/kJ, NO_2 was 8.4 μg/kJ, HCHO was 0.49 μg/kJ, and the submicron particulate emission rate was less than 0.04 μg/kJ. The shutter setting did not affect the CO and HCHO emissions of the 40C heater as much as it did those of the 30A heater. The burner design in heaters from manufacturer C is very different from that of the other two manufacturers. Instead of having a flat rectangular burner with many small circular ports which produce many small "flamelets," the 40C burner has relatively few slots cut across a cylindrically-shaped burner which produce softer "feathered" flames.

Field Test Results

The field emission rate test results of four UVGSHs (12A, 30A, 16B, and 30C) are described in Table II. Two of the heaters (30A and 16B) were used to assess the effects on indoor pollutant levels of varying air exchange rates.

Because steady state concentrations were not reached before the UVGSH was turned off, the final pollutant concentration is a fraction of the steady state concentration. That fraction was determined theoretically from Eq. 2 using the measured air exchange rate of

the house and the combustion time of the appliance. A "projected" steady state value was then determined by correcting the final whole-house concentration. At least 77% of the projected steady state concentration was reached in every experiment except for the two-hour experiments on heaters 30C and 12A, where approximately half of the projected steady state concentrations were reached. Typically, final indoor concentrations were over 90% of the steady state concentration. At the end of the UVGSH tests, the indoor/outdoor temperature differences ranged from 4.6°C (8.3°F) to 22.6°C (40.7°F).

During the field tests, the pollutants mixed rapidly throughout the house, presumably because of the large convective forces of the heater and the lack of barriers in the house—all interior doors were open. At the end of the UVGSH burns, CO_2 concentrations in the kitchen, living room, and bedroom were within 14% of their mean for all tests and, on average, were within 8% of their mean. Even when the heater was operated in the bedroom, rapid mixing took place. The attainment of uniform temperature in various parts of the house was generally not as rapid as the attainment of uniform pollutant concentrations and is probably explained by the thermal storage capacity of the walls and other interior surfaces that buffer changes in air temperature.

Carbon dioxide and oxygen. A graph depicting the projected steady-state concentrations for CO_2 versus the air exchange rate of the research house is shown on Figure 2. The curved lines drawn on the figure represent empirical fits of the data to the reciprocal of the air exchange rate for the 30A/30C heaters and the 16B heater. These curves demonstrate that the steady state concentrations of nonreactive gases whose source strength is constant follow the theoretical dependence on air exchange rate. The curves, less background, are straight lines in log-log space with an intercept equal to $\ln (S/V)$. Because V (and R) are known, S (and E) can be calculated. For CO_2, E was calculated to be 51,600 μg/kJ for the 30A/30C heaters

and 53,500 μg/kJ for the 16B heater. These values are within 5% of both the laboratory-derived emission rate of 51,100 μg/kJ and the theoretically-derived emission rate of 51,000 μg/kJ.

The consumption rates of O_2 were derived from the data in a similar manner. The results showed O_2 consumption rates of 68,900 μg/kJ for the 30A/30C heaters and 65,200 μg/kJ for the 16A heater. These values are within 9% of the laboratory-derived O_2 consumption rate of 70,900 μg/kJ and within 11% of the theoretical O_2 consumption rate of 73,200 μg/kJ.

Although, at the levels observed, the depletion of O_2 would not normally be considered a health hazard, CO_2 levels did exceed existing air quality standards and guidelines. The U.S. Occupational Safety and Health Administration (OSHA) has promulgated a time-weighted average CO_2 occupational standard of 5000 ppm for an eight-hour exposure,[14] and the American Society of Heating, Refrigerating, and Air Conditioning Engineers, Inc. (ASHRAE) has recommended a CO_2 "continuous guideline" of 2500 ppm.[15] The steady state CO_2 levels generated from the 30A/30C heaters exceeded the ASHRAE guideline in every experiment and exceeded the OSHA standard for all experiments where the air exchange rate was below 0.8 ach. The steady state CO_2 levels generated by the 16B heater approached the OSHA standard at 0.46 ach and exceeded the ASHRAE standard for all experiments below 0.9 ach.

Carbon monoxide. The final observed CO concentrations (measured just be-
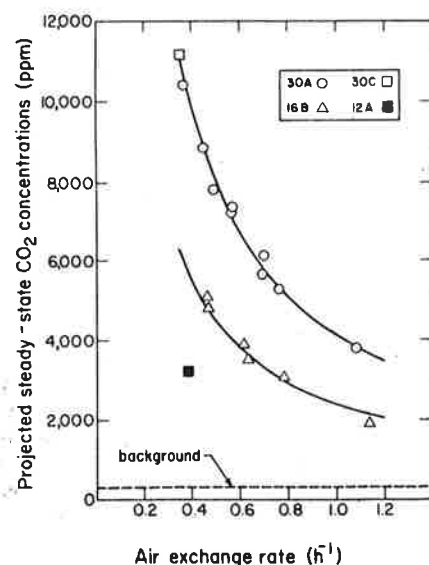
Figure 2. Projected steady state CO_2 concentrations associated with the use of unvented gas-fired space heaters vs. the air exchange rate of the house. Tests were conducted in a 240-m^3 unoccupied research house. The solid lines are empirical fits of the data to the reciprocal of the air exchange rate for the 30A/30C and 16B heaters.
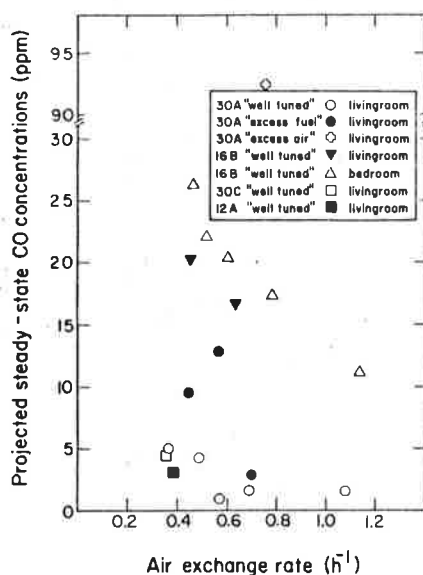
Figure 3. Projected steady state CO concentrations associated with the use of unvented gas-fired space heaters vs. the air exchange rate of the house. Tests were conducted in a 240-m³ unoccupied research house.

fore the heaters were turned off) ranged from 1.0 to 23 ppm, with the exception of one maltuned test, where it reached 89 ppm. The projected steady state concentrations are shown graphically on Figure 5. As observed in our laboratory studies, CO emission rates are not constant from heater to heater nor for an individual heater with different air to fuel ratios. For these reasons, we applied a different analysis for CO from that used for CO_2 and O_2.

One of the primary goals of this study was to investigate the applicability of laboratory-derived emission rates to field conditions. To accomplish this task, CO emission rates observed in the field were calculated using a constrained least-squares technique on the rise portion of the pollutant profile. The source strength, S, of CO was allowed to vary while the air exchange rate was constrained to be the value determined using CO_2 as the tracer, since at steady state the pollutant source strength and the air exchange rate cannot be simultaneously determined. Because we constrained the fitted initial concentration to be equal to the measured initial concentration, our procedure does not give as much weight to the early points (near ignition) as to the later points (near steady state). A list of these regression-derived "average" emission rates are included in Table II along with those for NO, NO_2 and N (of NO_x).

In addition to the "average" emission rates determined by regression, CO source strengths were estimated for the interval between every two whole-house CO data points, (i.e., every 24 minutes except for the initial estimate). These estimates were made in two steps. First,

two adjacent whole-house CO and CO_2 estimates were inserted into Eq. 3 as $C(0)$ and $C(T)$. (The other parameters in Eq. 3 were determined elsewhere.) This step yielded "uncorrected, semicontinuous" CO and CO_2 emission rate values at 24-minute intervals. Second, the uncorrected CO emission rate values were adjusted by multiplying them by the ratio of 51,100 $\mu g/kJ$ (the known CO_2 emission rate) and the uncorrected CO_2 emission rate. This second step increased the accuracy of our semicontinuous CO emission rates by reducing the effect of short-term incomplete mixing, which should affect CO_2 in the same manner as it affects CO.

The results of this semicontinuous analysis of source strength shows that CO emissions can radically change with time. The initial emission rate of CO, such as that measured in the laboratory, can be very different from the final steady state emission rate. Figure 4 shows the great variability in the temporal CO emission rate profile for representative tests. The CO emission rates during some heater tests drop after an initial warm-up period (see Figure 4). After the warm-up period, changing O_2, CO_2, and H_2O concentrations in the house air may also affect the temporal CO emission rate profile by slightly changing flame temperature or other flame characteristic. This may explain the rise in the CO emission rates for some tests between approximately 20 and 90 minutes after ignition.

Carbon monoxide emission rates can vary substantially from heater to heater, test to test, and also with time. While the reasons for the variation in CO
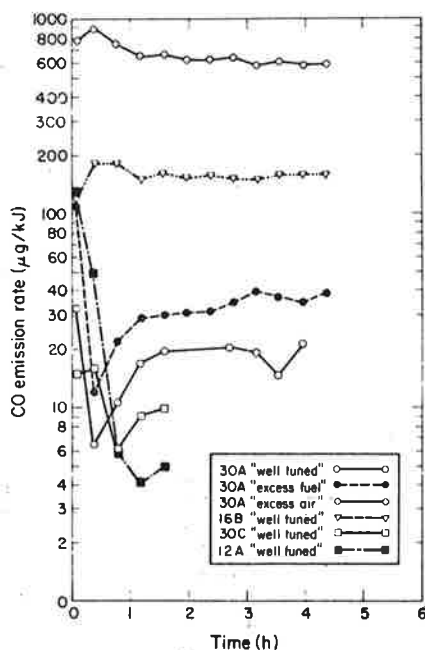
emission rates are not well understood, it is apparent that tuning (i.e., the primary air/fuel ratio) plays a major role in the variations observed for a single heater. It is also apparent that the CO levels in a house with an UVGSH can exceed outdoor air quality standards. Nine of the eighteen tests resulted in projected steady state CO levels exceeding the EPA's eight-hour standard of 9 ppm.[16] Seven of these tests involved the 16B heater. The 30A heater tested under excess-air (maltuned) conditions exceeded the EPA's one-hour standard of 35 ppm.
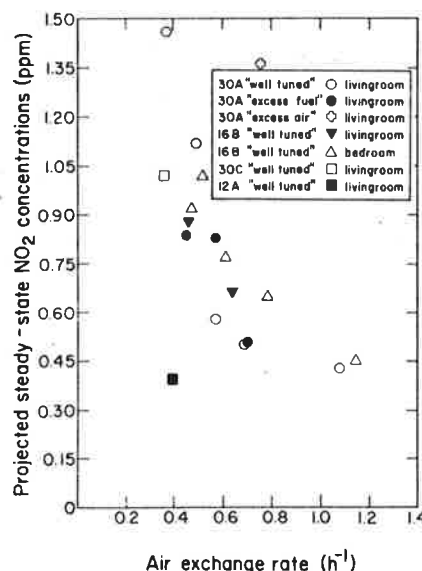
Figure 5. Projected steady state NO_2 concentrations associated with the use of unvented gas-fired space heaters vs. the air exchange rate of the house. Tests were conducted in a 240-m³ unoccupied research house.

Nitrogen oxides. Measured NO, NO_2 and NO_x levels at the time the heater was shut off ranged from less than 0.01 to 5.1 ppm for NO; 0.23–1.4 ppm for NO_2; and 1.2–6.4 ppm for NO_x. Average field emission rates and semicontinuous emission rates were calculated for NO, NO_2, and N (of NO_x). These rates were calculated as reported with CO, except that a reactivity rate term was included in the model.

The reactive decay rate, as defined by our model, is the net first-order rate of removal of a pollutant by means other than air exchange. In other words, the difference between the decay rate of a reactive pollutant and that of a nonreactive pollutant, such as CO_2, after the source is turned off yields the first-order reactivity rate. The reactivity rates measured during the pollutant decay periods were applied throughout the test. The average reactivity rates for NO and NO_x, 0.04 $\pm$ 0.08 h^{-1} and 0.08 $\pm$ 0.08 h^{-1}, respectively, are similar to those observed previously[5] in the same research house where a gas-fired range

Figure 4. CO emission rates vs. time for representative controlled field tests in an unoccupied research house.

was the pollutant source. However, the NO_2 reactivity rates were much lower, averaging 0.20 ± 0.13 h^{-1} in the present tests in contrast to 1.29 ± 0.67 h^{-1} in the earlier tests. The major differences between the two studies are that water was boiled during the earlier tests and the interior of the house had been painted between the two studies. However, there was no significant difference in the whole-house average relative humidities observed in the two studies.

By using the reactivity rates along with the air exchange rates, it was possible to determine how close our final measurement of whole-house pollutant concentrations was to our projected steady-state levels. The NO_2 levels for the 30A and 16B heater tests reached between 83.4% and 99.9% of steady state. Projected steady state levels were calculated for NO_2 and are shown in Figure 5.

Semicontinuous emission rate data for representative tests are shown graphically in Figure 6 for NO_2 and Figure 7 for N (of NO_x). For N (of NO_x), the emission rate consistently increases during an initial warm-up period, peaks between approximately 15 and 60 minutes of operation and then gradually decreases. After approximately 90 minutes, the N (of NO_x) emission rate appears fairly stable. Although not all the factors that affect the temporal N (of NO_x) emission rate profile are known, it appears to be affected by at least two phenomena. First, the N (of NO_x) emission rate rapidly increases while the heater is warming up and, presumably, the flame temperature is increasing. Second, the N (of NO_x) emission rate decreases as the O_2 level decreases and the CO_2, H_2O, and other combustion product concentrations increase. This
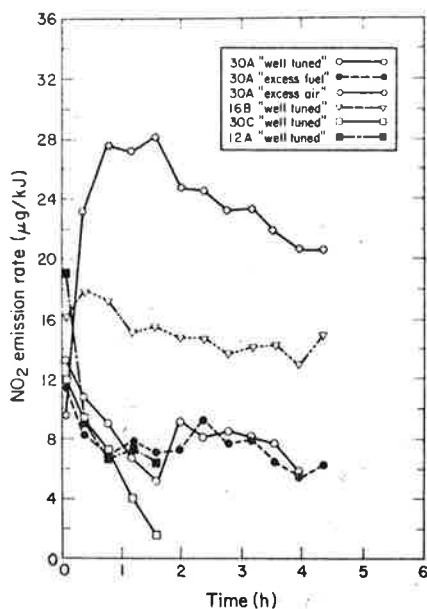

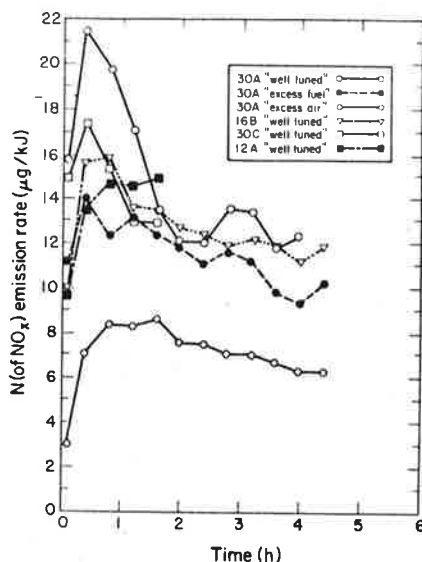Figure 6. NO_2 emission rates vs. time for representative controlled field tests in an unoccupied research house.

Figure 7. N (of NO_x) emission rates versus time for representative controlled field tests in an unoccupied research house.

change in supply air composition may cause cooling of the flame, thus accounting for the general decrease in the N (of NO_x) emission rate. Although consistent with the temporal CO emission rate profile of some tests, this explanation is speculative and further research is needed to test this hypothesis. The NO_2 temporal emission rate profiles are more complicated than those of N (of NO_x), probably because the NO_2 emission depends on factors other than flame temperature while the N (of NO_x) emission rate is driven primarily by the flame temperature.

Of NO, NO_2, and NO_x, only NO_2 is regulated by outdoor standards. The only short-term NO_2 standard promulgated by a state or federal organization is the state of California standard of 0.25 ppm for a one-hour average exposure.[17] All projected NO_2 steady state concentrations and all but one of the actual final NO_2 concentrations exceeded this one-hour standard (see Figure 5). The single exception for the final NO_2 concentration was for a two-hour test with heater 12A with a fuel consumption rate of only 9510 kJ/h. The final NO_2 level reached in that test was 0.23 ppm.

Particulates and formaldehyde. In general the particulate and formaldehyde concentrations were low. The highest sustained submicron particulate concentration was approximately 40 $\mu g/m^3$ during one test using the 30A heater under well-tuned conditions. This value is below the long-term EPA outdoor standard of 75 $\mu g/m^3$ for total suspended particulates.[16] However, it is not clear how applicable this standard is to indoor suspended particles since the chemical and physical nature of such particles can be very different from those of outdoor origin.

Of all the tests for formaldehyde, only one (heater 30A under excess air condition) showed formaldehyde levels to be above the most stringent HCHO standard, 0.10 ppm.[18] A concentration of 1.3 ppm was reached during that test. This test also produced high CO levels and was the only test conducted under the maltuned condition of excess air. The HCHO emission rate in this test was estimated to be 14 $\mu g/kJ$, assuming a reactivity rate of 0.4 h^{-1} based on earlier chamber results.[8]

Conclusions

The goals of this study were to quantify pollutant emission rates in the laboratory, to determine whether laboratory-derived emission rates were applicable to field situations, and to determine actual indoor pollutant levels caused by the use of an unvented gas-fired space heater in a real house. Laboratory-derived emission rates have been measured and can be correlated to initial field-derived emission rates. Laboratory data are also applicable when emission rates are constant over time such as for CO_2, O_2, NO_x and, for certain heaters, NO, NO_2, and CO. This study also confirmed the importance of appliance tuning especially for CO, NO_2, and HCHO emissions.

With regard to the third goal, we observed levels of CO_2 and NO_2 at or above existing standards or guidelines during most controlled field tests. Based on NO_2 findings alone, we must conclude that UVGSHs may pose a threat to the health of occupants living in houses where such appliances are used. Furthermore, depending upon such factors as the size of the heater, its state of tune, the volume and air exchange rate of the house, and the occupants' heater usage pattern, exposure to CO_2, CO, and HCHO may also exceed concentration limits set by environmental health or government agencies and may pose an additional health hazard.

views of the U.S. Consumer Product Safety Commission or the U.S. Department of Energy.

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