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G.W. Traynor, J.R. Allen, M.G. Apte, A.R. Carruthers, J.F. Dillworth, J.R. Girman, and V.M. Martin

May 1983

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ENERGY AND ENVIRONMENT DIVISION

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Indoor Air Pollution Due to Emissions from Unvented Gas-fired Space Heaters: A Controlled Field Study

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May 1983

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Abstract

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Operation of an unvented combustion appliance indoors can elevate pollutant concentrations. Indoor air pollution caused by using unvented gas-fired space heaters (UVGSHs) was investigated under controlled field conditions. Four UVGSHs were tested in a $240 - m^3$ research house with 0.36 to 1.14 air changes per hour. Based on measurements near steady state, we projected steady-state pollutant and oxygen levels: 1930 to 11,100 ppm for CO_2 ; 1.0 to 26 ppm for CO (for well-tuned heaters); 0.40 to 1.46 ppm for NO_2 ; and 19.1 to 20.7% for O_2 . Concentrations of CO_2 , CO, and $NO₂$ were sometimes observed to be above their outdoor or occupational guidelines. Analysis showed that CO , NO , and $NO₂$ emission rates can vary with time and that while short-term emission rates derived from earlier laboratory tests were consistent with initial emission rates observed in the field, they did not always correspond to steady-state emission rates.

Keywords: air shutter, combustion, carbon dioxide, carbon monoxide, emission rates, formaldehyde, indoor air pollution, nitric oxide, nitrogen dioxide, nitrogen oxides, pollutants, submicron particles, space heater, tuning, unvented.

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INTRODUCTION

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It has been demonstrated that operating unvented combustion appliances indoors increases the indoor concentration of several pollutants.¹⁻⁵ One such appliance currently of interest to the U.S. Consumer Product Safety Commission and the U.S. Department of Energy is the unvented gas-fired space heater (UVGSH). The indoor pollutant levels resulting from the use of UVGSHs depend on a wide range of factors including appliance type, appliance tuning, usage patterns, and building parameters (e.g., volume and air exchange rate). The two goals of this phase of our study were to determine if earlier short-term emission rates derived from the laboratory phase of this project would be applicable to field situations and to measure pollutant levels from the use of UVGSHs in a residential setting.

Earlier laboratory tests of UVGSHs conducted by Girman et al. measured the pollutant emission rates from eight UVGSHs and conducted extensive tests on the effects of tuning these UVGSHs.⁶ The laboratory results showed that UVGSHs emit carbon dioxide $(CO₂)$, carbon monoxide (CO), nitrogen dioxide (NO₂), nitric oxide (NO), formaldehyde (HCHO), and respirable particles. In addition, operation of UVGSHs reduced indoor oxygen ($0₂$) levels. Predictions applying laboratory emission rate data to projected conditions of use showed that levels of CO , $CO₂$, NO₂, and HCHO could exceed outdoor or occupational air quality standards of state or federal agencies or guidelines of scientific organizations. All the pollutant emission rates, except for CO_2 , were dependent on the tuning of the UVGSH. The tuning or "state-of-tune" of an UVGSH is adjusted with a simple butterfly valve located at one end of the burner assembly. The butterfly valve controls the air/fuel ratio of the appliance, which in turn controls the pollutant emission characteristics of the flame.

This field study of indoor air pollution from UVGSHs is a continuation of the laboratory study.⁶ The laboratory study investigated the effects of appliance type, fuel consumption rate, and tuning on pollutant emission rates; this field study primarily investigated the applicability of using short-term laboratory-derived emission rates and an

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indoor air quality model to predict indoor pollutant levels in an actual residence. In addition, the effects of air exchange rate, appliance tuning, and duration of combustion were investigated under controlled field conditions.

EXPERIMENTAL

For the field tests, four representative unvented gas-fired space heaters were selected from the eight tested in the laboratory phase. The heaters were purchased in 1981; they were not equipped with oxygendepletion sensors. All heaters burned natural gas that was similar in composition to the natural gas used in the laboratory study. The fuel ratings of the heaters used in this study ranged from 12,000 Btu/h (12,600 kJ/h) to 30,000 Btu/h (31,600 kJ/h).

All measurements were made at an unoccupied $107-m^2$ (1150-ft²) onestory experimental research house having an air volume of 240 m^3 . Figure 1 depicts the floor plan of the house as well as the air quality sampling sites and heater locations. 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 was well defined. No mixing fans were used. Air exchange rates, typical of the U.S. housing stock, were obtained by opening doors and windows from 0.5 em to 3.2 em, creating an additional leakage area in the building envelope between 400 cm^2 and 1400 cm^2 and a total air exchange rate between 0.36 h⁻¹ and 1.14 h⁻¹. All interior doors to rooms were open during all field tests.

Most measurements were made with the Lawrence Berkeley Laboratory's Mobile Atmospheric Research Laboratory (MARL), which is capable of remote multipoint sampling of CO_2 , CO , NO_2 , NO , O_2 , temperature, dew point, and wind speed (see Figure 2). All MARL data were initially recorded at one-minute intervals. CO_2 , CO_2 , NO_2 , NO_3 and O_2 concentrations in the outdoor, kitchen, living room, and bedroom air were measured on a rotating basis. Each site was monitored for six minutes. Data generated during this rotating cycle were analyzed by discarding the first three points and averaging the last three data points to obtain one observation every six minutes, i.e., 24 minutes elapsed between observations at each site. To eliminate sample-line purge time, air was

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continuously drawn into the MARL from each sampling site. The air drawn from the house into the MARL increased the air exchange rate by approximately 0.01 h⁻¹. Temperature and dew point, measured at each of the four locations, and wind speed, measured at the top of the house, were recorded every minute. A more complete description of MARL is contained in the report on laboratory investigations.⁶

Formaldehyde and submicron particulate measurements were made independently of MARL. Formaldehyde levels were measured using a refrigerated bubbler sampler to collect the formaldehyde for subsequent laboratory analysis.⁷ Sampling times were one or two hours and, in most tests, one sample was taken near the time the appliance was turned off, when the pollutant concentrations were high. The concentration of suspended particles below $0.5 \mu m$ in diameter were measured using an electrical mobility analyzer. 8 Typically, particulate measurements, made in the living room only, were recorded every 10 minutes.

A UVGSH was operated in either the living room or the bedroom as shown in Figure 1. The fuel consumption rate was measured with a diaphragm gas meter directly upstream of the heater. Fuel line pressure was set within manufacturers' specifications. As in laboratory tests, precautions were taken to prevent the heater's pilot light from burning until the main burner was lit. A piezoelectric sparker was used to ignite the burner. In each experiment, background indoor and outdoor pollutant concentrations were measured for approximately one hour before turning on the UVGSH to be tested.

Modeling

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Throughout the text, references will be made to a single-equation indoor air quality model based on mass balance. This model has been successfully used by many researchers for predicting indoor air pollution levels, as well as for determining indoor air quality parameters, such as pollutant emission rates, which can affect such levels.⁵,6,9-11 The model is repeated here for reference. The reader is referred to our earlier laboratory report on UVGSHs for more information.⁰

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The mathematical expression for the change in whole-house average indoor pollutant concentration is:

$$
dC = PaC_0 dt + \frac{S}{V} dt - (a + k) C dt,
$$
 (1)

where:

- $C =$ indoor pollutant concentration (ppm);
- $P = fraction of the outdoor pollutant concentration that$ penetrates the building shell (unitless);
- a = air exchange rate in air changes per hour (ach) (h^{-1}) ;
- C_{Ω} = outdoor pollutant concentration (ppm);
	- $t = time (h);$
	- $s =$ indoor pollutant source strength $(cm³/h)$;
	- $V = volume(m^3);$ and
	- k = net rate of removal process other than air exchange (h^{-1}) .

For particles, C and C_o are in units of μ g/m³ and S is in units of μ g/h. Assuming C_0 , P, a, S, and k are constant over the time period of interest, Eq. 1 can be solved for C(t) to give:

$$
C(t) = \frac{PaC_0 + S/V}{a + k} \left[1 - e^{-(a + k)t} \right] + C(0) e^{-(a + k)t}.
$$
\n(2)

The pollutant emission rate E (μ g/kJ for particles and cm^3/kJ for gases) of an appliance can be determined by solving Eq. 2 for S, dividing it by the fuel consumption rate R (kJ/h), and letting T equal the duration of appliance operation:

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

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For gases, E in cm^3/kJ can be converted to μ g/kJ by using the ideal gas law. For calculations involving the above equations, the penetration factor P was assumed to be 1.0 for gases and 0.4 for particles.¹¹

Because average concentrations of the entire house are required for the model and for the determination of air exchange rates and pollutant reactivity, this report frequently gives averages of the gas-phase pollutant data from the three indoor locations (kitchen, living room, and

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bedroom). This technique produces an estimate of the whole-house average gaseous pollutant concentration every 24 minutes.

The air exchange rate of the house was determined for each test by taking a time-weighted average of the air exchange rate computed during both the rise and fall of the $CO₂$ concentration. The "rise" air exchange rate was determined by using a generalized least-squares computer program to fit the $CO₂$ rise data to Eq. 2. For this procedure we set P equal to 1 and k equal to 0, and used the laboratory-derived source strength of CO_2 (51,100 μ g/kJ x fuel consumption rate). The "decay" air exchange rate was also determined from Eq. 2 using generalized least squares, but in this case the $CO₂$ source strength was zero. The difference between the decay and rise air exchange rates averaged 0.04 ± 0.10 h^{-1} , which is not significantly different from zero.

Test Description

Eighteen tests were conducted on four heaters. Two of the heaters were tested under several air exchange rates varying from 0.37 to 1.14 air changes per hour (ach). The average coefficient of variation of the rise and decay air exchange rates was 10%, which gives a combined estimate of the uncertainty in the air exchange rate measurements and/or the variation of the actual air exchange rates. A description of the basic parameters of each test is contained in Table 1. Each test is given an identification code that will be used throughout this report. The first two numbers followed by a letter represent the specific heater used in the study. The coding of the heaters is the same as that used in the laboratory report.⁶ The first two numbers, when multiplied by 1000, are the manufacturers' heat input rating in units of Btu/h (1.054 kJ/h); the letter represents one of the three heater manufacturers, coded as A, B, or C. The indoor/outdoor temperature difference when the UVGSH was turned off varied from 4.6 °C (8.3 °F) to 22.6 °C (40.7 °F), values that are typically encountered in real situations. Table 1 provides a convenient listing of the experiments but is not in chronological order.

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RESULTS AND DISCUSSION

Pollutant, temperature, dew point, and wind speed data are graphically represented in Appendix B.

During the field tests it was observed that the pollutants mixed rapidly throughout the house. This was presumably due to the convective forces of the heater. For example, just before the UVGSH was turned off, the kitchen, living room, and bedroom $CO₂$ concentrations were within 14% of their mean for all tests and, on average, were within 8%, lending validity to our use of whole-house average data throughout this report.

It is useful to discuss the field results by pollutant because various parameters affect individual pollutants differently (e.g., CO emissions are very sensitive to tuning while CO_2 emissions are not). It is also useful to compare the observed pollutant levels to outdoor and/or occupational guidelines because there are no promulgated residential air quality standards.

Carbon Dioxide and Oxygen

The final measured concentrations and estimated steady-state concentrations of CO_2 and O_2 are listed in Tables 2 and 3, respectively. The final concentrations represent the last whole-house measurement made before the UVGSH was turned off. All final concentrations for $CO₂$ and $0₂$, less background levels, were within 11% and 20%, respectively, of the theoretical values obtained using Eq. 2, the $CO₂$ and $O₂$ laboratoryderived emission/consumption rates, and the $\texttt{CO}_2\texttt{-}$ derived air exchange rate. On average, the final values were within 4% of the theoretical values for $CO₂$ and 7% for $O₂$.

In theory, steady-state concentrations are never reached. Therefore, for all tests, the final pollutant value was a percentage of the steady-state concentration. That percentage was theoretically determined by inserting the house air exchange rate, assumed to be constant throughout each test, and the combustion time into Eq. 2. A "projected" steady-state value was then determined from the final whole-house

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concentration. At least 77% of the projected steady state was reached in every experiment except for the two short, two-hour experiments (Test $#30C-1$, $#12A-1$).

Graphs of the projected steady-state concentrations for CO_2 and O_2 versus the house air exchange rate are shown in Figures 3 and 4, respectively. The curved lines drawn on Figures 3 and 4 represent empirical fits of the data to the reciprocal of the air exchange rate for the 30A/30C heaters and the 16B heater. The curves, less background, are straight lines in "log-log" space with an intercept equal to ln (S/V). Because V is known, S was calculated. For CO_2 , the emission rate (S \div fuel consumption rate) was calculated to be $51,600$ μ g/kJ for the 30A/30C heaters and $53,500$ μ g/kJ for the 16B heater. These values are within 5% of both the laboratory-derived emission rate of 51,100 µg/kJ and the theoretical emission rate, determined from the chemical composition of the natural gas, of 51,000 μ g/kJ.⁶

The $0₂$ consumption rates were analyzed from the data in Figure 4 in a similar manner. The results show $0₂$ consumption rates of 68,900 μ g/kJ for the 30A/30C heaters and $65,200 \text{ µg/kJ}$ for the 16A heater. These values are within 9% of the laboratory-derived $0₂$ consumption rate of 70,900 μ g/kJ and within 11% of the theoretical $0₂$ consumption rate of 73,200 μ g/kJ.⁶ The modeled concentrations of CO₂ and O₂ as well as CO, $NO₂$, and NO_x are shown in Appendix C.

Although the depletion of $0₂$ at the levels observed is not expected to be a health hazard, $CO₂$ levels from some tests exceeded existing air quality standards and guidelines. The U.S. Occupational Safety and Health Administration (OSHA) has promulgated a time-weighted average $CO₂$ occupational standard of 5000 ppm for an eight-hour exposure.¹² The American Society of Heating, Refrigerating, and Air Conditioning Engineers, Inc. (ASHRAE) has recommended a $CO₂$ "continuous guideline" of 2500 ppm.¹³ The steady-state $CO₂$ levels generated from the 30A/30C heaters are above the OSHA standard for all experiments below 0.8 ach and above the ASHRAE guideline for every experiment. The steady-state $CO₂$ levels generated from the 16B heater approached the OSHA standard at 0.46 ach and exceeded the ASHRAE standard for all experiments below 0.9

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Carbon Monoxide

ach.

The final measured CO concentrations were below 25 ppm, ranging from 1.0 to 23.2 ppm, with the exception of one test of a maltuned heater (test 30A-9), in which concentrations reached 89.4 ppm. Table 4 lists the final CO concentrations measured before the heaters were shut off and the projected steady-state concentrations. Projected steady-state concentrations were calculated in the same manner as for $CO₂$ and $O₂$ and are shown graphically in Figure 5.

As shown in our laboratory studies, carbon monoxide emission rates are not constant from heater to heater or for an individual heater if the air/fuel ratio changes. These facts necessitated a different analysis strategy for CO than that used for CO₂ and O₂.

One of the primary goals of this study was to investigate the applicability of short-term (10 to 30 minutes of combustion) laboratoryderived emission rates to field conditions in which heaters were operated for longer periods of time. To accomplish this task, "average" CO emission rates were calculated from the field data by using a constrained least-squares technique on the rising 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 $CO₂$ and listed in Table 1. It was necessary to constrain the air exchange rate because, at steady state, the generalized least-squares technique cannot simultaneously separate the pollutant source strength and the air exchange rate. Since we fix the first theoretical point to be equal to the measured initial concentration, our procedure gives less statistical weight to the early points than to the later points. Table 4 lists these "average" emission rates (also called regression-derived emission rates).

In addition to the "average" emission rates determined by regression, estimates of the CO source strength were made 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, the two

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sequential whole-house CO and $CO₂$ concentration estimates were inserted into Eq. 3 as C(O) and C(T). (The other parameters in Eq. 3 were determined previously.) This step yielded "uncorrected" CO and $CO₂$ "semicontinuous" 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 μ g/kJ, the known CO₂ emission rate, and the uncorrected $CO₂$ emission rate. This second step increased the accuracy of our semi-continuous CO emission rate by reducing the effect of incomplete mixing since CO_2 should mix in the same manner as CO . Incomplete mixing was a problem primarily that occurred before steady-state was approached.

The results of this semi-continuous source strength analysis are compiled in Appendix A and summarized in Table 4. Representative tests are shown graphically in Figure 6. The results show that CO emissions can change with time. The initial emission rate of CO can differ greatly from the regression-derived "average" emission rate or the final emission rate, defined as the average of the last three semi-continuous emission rate points that are within the last 72 to 85 minutes of combustion.

In general, the CO emission rate profiles were not monotonic (see tests 30A-2 and 30A-6 on Figure 6). This allows the possibility that the average CO emission rate is not found between the initial and final values. The factors that affect CO emission rates over time are not well understood. It appears that the CO emission rate during some heater tests drops after an initial warm-up period (see tests 30A-2, 30A-6, 30C-l, and 12A-1 in Figure 6). After the warm-up period, changing 0_2 , CO_2 , and H_2O concentrations may also affect the temporal CO emission rate profile by slightly changing flame temperature or other characteristics. This may explain the rise in the CO emission rate for the 30A-2, 30A-6, and 30C-l tests between approximately 20 and 90 minutes.

The regression-derived emission rates of the 30A heater under welltuned conditions ranged from 2.4 to 16 μ g/kJ. This is lower than the 25 fg/kJ measured in our laboratory tests with a combustion time of

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approximately 10 minutes. However, the average of the initial CO emission rates was 25 ± 6 µg/kJ, which agrees well with our laboratory test results.

The 30A "closed shutter" tests (30A-6, 30A-7, and 30A-8) showed dramatically lower field CO emission rates than did the laboratory test. The field rates varied from 18.8 to 57.7 μ g/kJ, whereas the laboratory rate was 159 µg/kJ. Part of this discrepancy can be attributed to the reduction of the CO emission rate with time, but it appears that, with the air shutter closed, the 30A heater emitted less CO in the field than in the laboratory.

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The regression-derived CO emission rate and initial CO emission rate of the 30A under the excess air condition (shutter 69% open) of 620 μ g/kJ and 780 μ g/kJ, respectively, were above the laboratory-derived emission rate of 517 µg/kJ under the fully open shutter condition. Although this comparison is not fully justified due to the different shutter settings, it illustrates that high CO emissions can be obtained at shutter openings greater than 69%.

The CO emission rates of the 16B heater were stable over time, especially when compared to those of the other heaters. The laboratory emission rate of $190 \mu g/kJ$ appears to agree more with the emission rates determined from the tests conducted in the bedroom (tests 16B-3 through 16B-7) than with the tests conducted in the living room (tests 16B-1 and 16B-2). The mean of the regression-derived emission rates for tests 16B-3 through 16B-7 was 200 \pm 10 μ g/kJ, and the mean of the initial emission rate values was 200 \pm 30 μ g/kJ, which illustrates this agreement.

The regression-derived CO emission rate of the 30C heater was 12 μ g/kJ; the initial rate was 15 μ g/kJ. For the 12A heater the initial CO emission rate of 130 µg/kJ was consistent with the laboratory rate of 114 µg/kJ. The "average" and final emission rate values were much lower, demonstrating the large change over time in the CO emission for this heater.

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Carbon monoxide emission rates can vary substantially from heater to heater, from test to test, and with time. The reasons for the variation in CO emission rates are not well understood, but clearly tuning (i.e., the air/fuel ratio) plays a major role in the variations observed within tests of a single heater. It is also apparent that the CO levels in a house with a UVGSH can exceed outdoor long-term and short-term standards. Nine of the 18 tests had projected steady-state CO levels that exceeded the Environmental Protection Agency's eight-hour standard of 9 ppm.¹⁴ Seven of these tests were conducted with the 16B heater. The EPA's one-hour standard was exceeded once when using the 30A heater under the excess air (maltuned) condition.

Nitrogen Oxides

The measured and calculated results for NO, NO_2 , and NO_x are reported in Tables 5, 6, and 7, respectively. Observed levels ranged from 0.00 to 5.14 ppm for NO; 0.23 to 1.35 ppm for NO₂; and 1.21 to 6.36 ppm for NO_{x} . To compare the pollutant levels observed in the field with those based on short-term (10 to 30 minutes of combustion) laboratory tests, it was necessary to derive "average" field emission rates and semi-continuous emission rates for NO, NO_2 , and N (of NO_x). This was accomplished in a manner similar to that used for CO, with the exception that a reactivity term was needed. The reactivity, as defined by our model, is the net first-order rate of removal of a pollutant by means other than air exchange. Therefore, the difference between the decay rate of a reactive pollutant and that of an unreactive pollutant (e.g., $CO₂$) after the source is turned off yields the first-order reactivity rate. It was assumed that reactivity rates measured during the pollutant decay periods applied throughout the test. listed in Tables 5, 6, and 7. Measured reactivities are

The average reactivity rates for NO and NO_x , 0.04 \pm 0.08 h⁻¹ and $0.08 \pm 0.08 \text{ h}^{-1}$, respectively, are similar to those observed by Traynor et $a1.^5$ in the same house using a gas-fired range as the pollutant source, but the $NO₂$ reactivities were much lower than those reported earlier. The average $NO₂$ reactivity of the tests in this report was 0.20 \pm 0.13 h⁻¹ whereas the earlier study reported 1.29 \pm 0.67 h⁻¹.

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Major differences between the two studies are that the earlier study boiled water during the test and that the interior of the house was painted between the two studies, suggesting that water vapor or the $NO₂$ deposition rate on painted surfaces may play a role in the overall $NO₂$ reactivity rate. However, there was not a major difference between the average relative humidity levels of the two studies.

Using calculated reactivity rates and the air exchange rate made it possible to determine how close our final whole-house concentration measurement was to steady state. The $NO₂$ levels for the 30A and 16B heater tests reached between 83.4% and 99.9% of steady state (see Table 6). Projected steady-state levels were calculated and are shown in Figure 7.

The semi-continuous emission rate data for NO, NO_2 , and N (of NO_x) are listed in Appendix A and summarized in Tables 5, 6, and 7. Selected tests are shown graphically in Figure 8 for $NO₂$ and Figure 9 for N (of For N (of NO_x), there is a consistent trend where the emission NO_{ν}). rate increases for an initial warm-up period, peaks between approximately 15 and 60 minutes of operation, then gradually decreases with time. 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_y) emission rate decreases as the $0₂$ level decreases and the CO_2 , H_2O , and other combustion product concentrations increase. This change in supply air composition may cause cooling of the flame, thus accounting for the general' decrease in the N (of NO_v) 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 such a hypothesis. The $NO₂$ temporal emission rate profiles are more complicated than those of NO_x , probably because the NO₂ emission depends on many factors while the NO_x emission rate is driven primarily by the flame temperature.

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The mean of the regression-derived emission rates for N (of NO_x) from the 30A heater under well-tuned conditions was 15.7 \pm 2.2 μ g/kJ, which is slightly above the laboratory rate of 13.6 µg/kJ. The initial emission rates for the same tests averaged 13.7 \pm 1.4 μ g/kJ, a value closer to the laboratory-derived rate. The mean of the regressionderived NO₂ emission rates for these tests was 7.5 \pm 1.9 µg/kJ, which was lower than the laboratory rate of 11.4 µg/kJ. However, the initial NO₂ emission rate, averaging 10.4 ± 3.0 µg/kJ, is more consistent with the laboratory rate.

Similar results were obtained from the excess-fuel tests with the 30A heater (tests 30A-6, 30A~7, 30A-8). The average initial emission rate for N (of NO_y), 11.1 \pm 0.3 µg/kJ, is closer to the laboratory rate of 11.2 µg/kJ than is the mean of the regression-derived emission rates, 12.8 \pm 0.7 µg/kJ. And the average initial NO₂ emission rate of 11.6 \pm 2.2 μ g/kJ is closer to the laboratory rate of 13.7 μ g/kJ than is the mean of the regression-derived emission rates, $9.0 \pm 1.7 \text{ µg/kJ}$.

When the 30A heater was operated with the air shutter 69% open, no increase in NO levels was observed; thus the emission rate for NO was below 0.1 μ g/kJ, which agrees with the laboratory-derived emission rate of 0.04 μ g/kJ when the heater air shutter was 100% open. The initial NO₂ and N (of NO_x) emission rates of 3.0 μ g/kJ and 9.7 μ g/kJ, respectively, were within 20% of the laboratory rates of 3.4 μ g/kJ for NO₂ and 11.5 μ g/kJ for N (of NO_x). However, the NO₂ and N (of NO_x) "average" and final emission rates were approximately twice the initial value.

The average initial NO, NO_2 , and N (of NO_x) emission rates for tests 16B-1 through 16B-7 were 16.3 ± 3.6 μ g/kJ, 18.3 ± 2.8 μ g/kJ, and 13.2 \pm 2.4 µg/kJ, respectively, and agreed with the laboratory-derived rates of 13.9 μ g/kJ, 18.1 μ g/kJ, and 12.3 μ g/kJ for NO, NO₂, and NO_x, respectively. The regression-derived NO emission rates averaged 20.4 ± 3.4 fg/kJ, approximately 25% higher than the initial emission rate. The regression-derived NO₂ emission rates averaged 17.8 \pm 1.9 µg/kJ, about the same as the initial values.

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Results from the 30C and 12A heater tests showed high initial $NO₂$ emission rates and low initial NO emission rates compared to the "average" rates. The laboratory-derived emission rates for NO and $NO₂$ emitted from the 30C heater were 19.3 pg/kJ and 9.6 pg/kJ, respectively, below initial NO and NO₂ emission rates of 24.1 μ g/kJ and 12.0 μ g/kJ, respectively. The laboratory-derived emission rates from the 12A heater were 9.6 μ g/kJ for NO and 19.7 μ g/kJ for NO₂, similar to the initial field emission rate values of 8.4 μ g/kJ for NO and 19.0 μ g/kJ for NO₂.

Of NO, NO_2 , and NO_x , only NO_2 is regulated by outdoor standards. The only outdoor short-term $NO₂$ standard promulgated by a state or federal agency is the California short-term standard of 0.25 ppm for a one-hour average.¹⁵ The projected NO_2 steady-state concentrations exceeded the California one-hour standard for every test (see Figure 7). All but one of the measured final $NO₂$ concentrations also exceeded the California short-term $NO₂$ standard. The lone exception was for test 12A-1, in which the UVGSH consumed only 9510 kJ/h and was operated for only two hours. The final $NO₂$ level reached in test 12A-1 was 0.23 ppm, just below the California short-term standard.

Particulates and Formaldehyde

Particulate and formaldehyde concentrations were generally low. The highest sustained submicron particulate concentration was approximately $4 \mu g/m^3$ during test 30A-1. This value is well below the long-term EPA outdoor standard of 75 μ g/m³ for total suspended particulates.¹⁴

Of all the tests in which formaldehyde was measured, only one (test 30A-9) had formaldehyde levels above the most stringent of formaldehyde standards, 0.10 ppm.¹⁶ A concentration of 1.3 ppm was reached during that test. It was also the test that produced high CO levels and the only test conducted with a heater under excess air (maltuned) conditions. An estimate of the HCHO emission rate was made by assuming a reactivity rate of $0.4 h^{-1}$ based on earlier chamber studies.¹¹ The field HCHO emission rate was $14 \text{ pg}/\text{kJ}$, which is lower than the $20.3 \text{ pg}/\text{kJ}$ measured in the laboratory with the shutter 100% open. There were insufficient data to calculate semi-continuous emission rates for HCHO. Since the shutter on test 30A-9 was only 69% open, the two values are

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consistent.

Additional Discussion

One of the more interesting observations made from this study was the very rapid mixing that occurred during all tests, as demonstrated by the similar pollutant concentrations observed at the kitchen, living room, and bedroom monitoring sites throughout the tests (see Appendix B). This even occurred during tests conducted in a bedroom with the door open. In general, the attainment of uniform temperature in various parts of the house was not as rapid. This is explained by the thermal storage in the walls and other interior surfaces that can buffer changes in temperature.

Because the air in the house mixed rapidly, our model's assumption of well-mixed air is a good one. On the other hand, our assumption that the source strength and emission rates from UVGSHs are constant is not always met. When the assumption of a constant source strength is met, the field and laboratory results are in good agreement. When the emission rates are not constant, the initial field emission rates usually agree with the laboratory-derived rates. The assumption of a constant source strength is appropriate for CO_2 and O_2 , but, in general, is less appropriate for CO, NO, NO_2 , and NO_x . However, the modeled final pollutant concentrations for. CO, NO, NO_2 , and NO_x using the regressionderived emission rates were, on average, within 10% of the measured value. The regression-derived emission rates, which yield average emission rates over an extended period of time, may be more useful than initial or final emission rates in the absence of information about consu~ mer usage patterns.

CONCLUSIONS

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The two primary goals of this study were to determine if laboratory-derived emission rates were applicable to a field situation and to determine actual indoor pollutant levels caused by using an unvented gas-fired space heater in a real house. With respect to the first goal, results showed that short-term laboratory emission rates are, in general, applicable to initial field emission rates. However,

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they are not generally representative of long-term emission rates except when the emission rates are constant over time, such as for CO_2 and O_2 . For some heaters, under certain tuning conditions, CO , NO , and $NO₂$ may also be reasonably constant. For those conditions under which NO_2 , NO_2 , NO_y, and CO emission rates are not constant with time, the use pattern of the UVGSH becomes an important factor in determining pollutant concentrations. However, using a single long-term emission rate value can adequately characterize pollutant concentrations near steady state. This study also confirmed the importance of the effect of tuning on emission rates. This holds true especially for CO , $NO₂$, and HCHO.

With regard to the second goal, we observed levels of $CO₂$ and NO₂ at or above existing standards or guidelines during most controlled field tests. $NO₂$ levels were consistently higher than the California one-hour outdoor \overline{NO}_2 standard (0.25 ppm).¹⁵ CO levels were often observed to be above over the EPA eight-hour standard (9 ppm)¹⁴ under well-tuned conditions and once were observed to be over the EPA one-hour standard (35 ppm)¹⁴ under a maltuned condition. CO_2 levels were above the OSHA occupational eight-hour standard $(5000 \text{ ppm})^{12}$ when the 31,600 kJ/h $(30,000$ Btu/h) heaters were operated with a ventilation rate less than 0.8 ach. HCHO levels greatly exceeded the American Industrial Hygiene Association's guideline $(0.10 \text{ ppm})^{16}$ during one maltuned test, but were low in all other cases. The lowest projected steady-state $0₂$ level during the field tests was 19.08%; all measured levels were above 19.48%.

Based on $NO₂$ findings alone, we must conclude that UVGSHs may pose a potential threat to the health of occupants of houses where such appliances are used. Depending on such factors as heater size, state of tune, house volume, house air exchange rate, and heater use pattern, occupant exposure to other pollutants such as CO_2 , CO , and HCHO may also exceed current recognized health standards and may pose an additional health hazard.

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Table 1. Description of Controlled Field Tests.

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 $a_{\text{Heat content of natural gas used} \approx 31.4 \text{ kJ/L.}$

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bAverage of kitchen, living room, and bedroom values minus the outdoor value; computed at end of combustion, Table 2. Carbon Dioxide Results from Controlled Field Tests.

aTime from ignition to last whole-house average concentration during combustion.

 b Last whole-house average concentration during combustion. Average background $C0₂$ concentration</sup> for all tests was 320 ± 30 ppm.

 $c(1 - e^{-at})$ x 100 where a is air exchange rate and t is elapsed

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combustion time.

d_{Calculated} concentration at time of last whole-house average concentration during combustion using indoor air quality model; used ω_2 emission rate of 51,100 μ g/kJ (laboratory average). e Error of modeled final CO_2 concentration compared with measured final CO_2 concentration.

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Background (outdoor) level was subtracted before error calculations were made.

Table 3. Oxygen Results from Controlled Field Tests.

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aTime from ignition to last whole-house average concentration during combustion.

 b Last whole-house average concentration during combustion. Average background $0₂$ concentrations</sup> for all tests was $20.92\% \pm 0.05\%$.

 $c(1 - e^{-at}) \times 100$.

d_{Calculated} concentration at time of last whole-house average concentration during combustion using indoor air quality model; used $0₂$ consumption rate of 70,900 μ g/kJ (laboratory average). ^e Error of modeled final $0₂$ concentration compared with measured final $0₂$ concentration. Background (outdoor) level was subtracted before error calculations were made.

aTime from ignition to last whole-house average concentration during combustion.

b_{Last whole-house} average concentration during combustion. Average background CO concentration

is 0.40 ± 0.24 ppm.

 $c_{(1-e^{-at})} \times 100$.

 d Determined by generalized least squares.

eFirst emission rate value within S to 11 minutes of ignition.

^fTime-weighted average of three emission rate values within 72 to 85 minutes of end of combustion.

gCalculated concentration at time of last whole-house average concentration during combustion

using indoor air quality model; used "average" CO emission rate listed on this table.

h_{Error} of modeled final CO concentrations compared with measured final CO concentration.

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Table 5. Nitric Oxide Results from Controlled Field Tests.

 $a_{\text{Mean}} = 0.04 \pm 0.08 h^{-1}$.

b_{Time} from ignition to last whole-house average concentration during combustion.

CLast whole-house average concentration during combustion. Average background concentration = 0.01 ± 0.01 ppm.

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d_{(1 - e^{-(a + k)t}) x 100}
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eDetermined by generalized least squares.

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fFirst emission rate value within 5 to 11 minutes of ignition.

 $g_{\text{Time}-\text{weighted}}$ average of three emission rate values within 72 to 85 minutes of end of combustion.

 h Calculated concentration at time of last whole-house average concentration during combustion using indoor air quality model;</sup> used "average" NO emission rate listed on this table.

iError of modeled final NO concentration compared with measured final NO concentration.

Table 6. Nitrogen Dioxide Results from Controlled Field Tests,

 $a_{\text{Mean}} = 0.20 \pm 0.18 h^{-1}$.

b_{Time} from ignition to last whole-house average concentration during combustion.

 c Last whole-house average concentration during combustion. Average background concentration = 0.03 \pm 0.01 ppm.

 $d_{(1 - e^{(a + k)t}) x 100}$.

eDetermined by generalized least squares.

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fFirst emission rate value within *5* to 11 minutes of ignition.

8Time-weighted average of three emission rate values within 72 to 85 minutes of end of combustion.

hcalculated concentration at time of last whole-house average concentration during combustion using indoor air quality model;

used "average" $NO₂$ emission rate listed on this table.

¹Error of modeled final NO₂ concentration compared with measured final NO₂ concentration.

Table 7. Nitrogen Oxides Results from Controlled Field Tests.

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 $a_{\text{Mean}} = 0.08 \pm 0.08 h^{-1}$.

 b Time from ignition to last whole-house average concentration during combustion.</sup>

 c Last whole-house average concentration during combustion. Average background concentration = 0.04 \pm 0.01 ppm. $d_{(1 - e^{-(a + k)t}) x 100}$.

eDetermined by generalized least squares.

fFirst emission rate value within 5 to 11 minutes of ignition.

&Time-weighted average of three emission rate values within 72 to 85 minutes of the end of combustion.

hcalculated concentration at time of last whole-house average concentration during combustion using indoor

air quality model; used "average" N (of NO_x) emission rate listed on this table.

¹Error of modeled final NO_x compared with measured final NO_x concentration.

Particulate Analyzer

Heater Location

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Figure 1. Schematic of the field research house.

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Figure 2. Schematic of the instrumentation used for controlled field experiments.

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Figure 3. Projected steady-state CO_2 concentrations caused by using
unvented gas-fired space heaters versus the house air unvented gas-fired space heaters versus the house air exchange rate. Tests were conducted in a $240-m^3$ unoccupied exchange rate. Tests were conducted in a 240-m³ 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.

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Figure 4. Projected steady-state $0₂$ concentrations caused by using unvented gas-fired space heaters versus the house air
exchange rate. Tests were conducted in a $240\neg m^3$ unoccupied exchange rate. Tests were conducted in a $240-m^3$ 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.

XBL 834-9042

Figure 5. Projected steady-state CO concentrations caused by using unvented gas-fired space heaters versus the house air exchange rate. Tests were conducted in a $240 - m³$ unoccupied research house.

Figure 7. Projected steady-state $NO₂$ concentrations caused by using
unvented gas-fired space heaters versus the house air unvented gas-fired space heaters versus the house air
exchange rate. Tests were conducted in a $240-m^3$ unoccupied exchange rate. Tests were conducted in a $240 - m³$ research house.

 $NO₂$ emission rates versus time for representative controlled field tests. Figure 8.

Figure 9. N (of NO_x) emission rates versus time for representative controlled field tests.

Semi-continuous emission rate data for CO , NO , $NO₂$, and NO_x .

 $(Cont.)$

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*High outdoor (background) concentration relative to indoor concentration invalidated emission rate value.

APPENDIX B

Appendix B graphically summarizes the pollutant, temperature, dew point, and wind speed data for all controlled field experiments. Pollutant data within a single 1ocation are 24 minutes apart. Temperature and dew point data represent 10-minute moving averages. Wind speed data represent 3Q-minute moving averages.

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LEGEND :

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OUTSIDE

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TEST: 30A-9 TEMPERATURE CO NO $\frac{1}{\sqrt{2}}$ OFF OFF n٨ 24 4.8 45 ក្ដិ៍ រ δ $\bar{\tilde{\mathbf{a}}}$ 3.2 း မွ³⁰ \pmb{a} $\mathbf{1.6}$ ${\bf 15}$ $\pmb{\mathsf{o}}$ 0.0 $\pmb{\mathfrak{g}}$ **NO2** DEWPOINT $CO₂$ $\overline{\mathbf{3}}$ 7500 30 $rac{6}{9}$ 5000 $\frac{e}{2}$? ၊ ပွ²⁰း 2500 \mathbf{I} $10\,$ $\mathbf 0$ \bullet $\pmb{\mathfrak{o}}$ WIND SPEED 02 **NOX** \equiv 20.8 $\pmb{6}$ 24 km/hr $\frac{5}{9}$ 4 $88.20.2$ 16 19.6 $\overline{2}$ $\pmb{\theta}$ 19.0 \bullet $\ddot{\mathbf{0}}$ 6 7 8
HOURS 8 10 11 12 13 PARTICLES
(<0.5µM) $0 \quad 1 \quad 2 \quad 3$ $4₅$ HCHO 120 $\mathbf 6$ LEGEND : $\frac{e \omega \tau}{\tau}$ **LIVINGROOM** $\frac{\Omega}{\Omega}$ 80 KITCHEN **BEDROOM** 40 \mathbf{z} - WHOLE-HOUSE OUTSIDE $\mathbf 0$ $\mathbf{0}$ 0 1 2 3 4 5 6 7 8 9 10 11 12 13
HOURS $6 \quad 7 \quad 6$ 9 10 11 12 13 **a** $\overline{\mathbf{5}}$ -2 \blacktriangleleft **HOURS** XBL 8210-4786

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TEST:16B-6 TEMPERATURE CO N₀ .
AEC ner 45 24 4.8 $\frac{\epsilon}{\Delta}$ 18 $\mathop{\mathsf{a}}\limits^{\mathsf{e}}_{\Delta^{3,2}}$ ္မွ ³⁰ 1.6 15 θ \bullet 0.0 \mathbf{a} DEWPOINT c_{02} N02 $\mathbf 3$ 30 7500 $\frac{2}{9}$ 5000 គ្និ ខ្ ^t පු ²⁰ 10 2500 \mathbf{I} \bullet \bullet \mathbf{o} WIND SPEED 02 **NOX** 20.8 24 $\,$ 6 $\,$ km/hr $\frac{\epsilon}{\alpha}$ 8820.2 19.6 \overline{c} \mathbf{a} 19.0 $\pmb{\mathfrak{o}}$ \bullet 0 1 2 3 4 5 6 7
HOURS 8 9 10 11 12 13 $\frac{PARTICLES}{(0.5 \mu M)}$ HCHO 120 $\bf{6}$ LEGEND : $\frac{4.9}{4}$ LIVINGROOM $\frac{5}{6}$ 80 KITCHEN **BEDROOM** 40 \overline{c} WHOLE-HOUSE OUTSIDE $\mathbf 0$ 0 1 2 3 4 5 6 7 8 9 10 11 12 13 10 11 12 13 0 1 2 3 $4₅$ $6₇$ θ 9 HOURS **HOURS** XBL 8210-4802

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ក្នុង **ខ** $\frac{\epsilon}{\alpha}$ 3.2 . ൃ³⁰. \mathbf{s} 1.6 15 0.0 \bullet $\mathbf 0$ $CO₂$ **NO2** DEWPOINT $\overline{\mathbf{a}}$ 7500 30 $\frac{6}{9}$ 5000 $\frac{\epsilon}{\Delta}$ 2 1 **ပွ**း 2500 $\overline{\mathbf{1}}$ 10 $\mathbf 0$ $\pmb{\mathfrak{o}}$ $\pmb{\mathfrak{o}}$ **NOX** WIND SPEED $02\,$ 20.8 \mathbf{s} 24 km/hr $\frac{\epsilon}{2}$ 4 20.2 18 $19.6\downarrow$ $\overline{2}$ \bullet $\mathbf{0}$ 19.0 \bullet 0 1 2 3 4 5 6 7 6 9 10 11 12 13
HOURS PARTICLES.
(<0.5µM) HCHO 120 $\pmb{6}$ LEGEND : $\frac{1}{2}$ LIVINGROOM $\frac{10}{10}$ 80 KITCHEN **BEDROOM** 40 $\mathbf{2}$ WHOLE-HOUSE OUTSIDE $\pmb{0}$ \bullet 0 1 2 3 4 5 8 7 8 9 10 11 12 13 0 1 2 3 4 5 6 7 8 9 10 11 12 13 HOURS HOURS XBL 8210-4803

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APPENDIX C

"Theoretical" and measured whole-house CO , O_2 , NO_2 , and NO_x concentrations for all controlled field tests. The theoretical concentrations were derived using a single-equation indoor air quality model (see Eq. 2), with some parameters derived from measured data. The air exchange rate was derived from the CO_2 data. The CO_2 and O_2 emission rates were taken from an earlier laboratory study. An "average" emission rate, determined by generalized least squares, was used for CO , NO_2 , and NO_x .

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