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INDOOR AIR POLLUTION DUE TO EMISSIONS FROM UNVENTED GAS-FIRED SPACE HEATERS

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April 1983

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#### Abstract

Operation of an unvented combustion appliance indoors can elevate pollutant concentrations. Under laboratory conditions, we determined the oxygen consumption rates and the pollutant emission rates of  $CO$ ,  $CO<sub>2</sub>$ , NO,  $NO<sub>2</sub>$ , HCHO and submicron particles emitted from eight unvented gas-fired space heaters operated with well adjusted air shutters at partial and full input in a  $27 - m<sup>3</sup>$ 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<sup>3</sup> 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:  $1,930$  to  $11,100$  ppm for  $CO_2$ , 1.0 to 26 ppm for  $CO$  (under well-tuned conditions), 0.40 to 1.46 ppm for  $\text{NO}_2$ , and 19.1 to 20.7% for  $\text{O}_2$ . Concentra tions of CO<sub>2</sub>, CO, and NO<sub>2</sub> were sometimes observed to be above their outdoor or occupationa $\widehat{\mathbb{1}}$  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.

#### 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.

#### Introduction

It has been demonstrated that operating unvented combustion appliances indoors increases the indoor concentrations of several pollutants. <sup>1-5</sup> One such appliance of current interest to the U.S. Consumer Product Safety Commission (CPSC) and the U.S. Department of Energy (DOE) is the unvented natural gas-fired space heater (UVGSH). This study examines the pollutant emissions from eight UVGSHs under laboratory conditions. In addition, four of the heaters were examined for their emissions and impact on indoor air quality under controlled field conditions. The eight UVGSHs tested in this report were purchased in 1981 from three U.S. manufacturers. They 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 fuel input ratings of the heaters ranged from 12,600 kJ/h (12,000 Btu/h) to 42,200 kJ/h (40,000 Btu/h).

The three basic goals of this study were to quantify pollutant emission rates in the laboratory, to determine if laboratory-derived emission rates were applicable to a field situation, and to determine actual indoor pollutant levels due to the use of an unvented gas-fired space heater in a residence. The indoor pollutant levels resulting from the use of UVGSHs depend on a wide range of factors including the appliance pollutant emission rates, appliance usage patterns, and building parameters (e.g., volumes and air exchange rates). Under laboratory conditions, all eight heaters were tested in an environmental chamber for their consumption of oxygen  $(0, 0)$  and for their emissions of carbon monoxide (CO), carbon dioxide (CO<sub>2</sub>), nitric oxide (NO), nitrogen dioxide  $(NO<sub>2</sub>)$ , formaldehyde (HCHO), and submicron particles. All of the heaters were tested under well-tuned conditions and some heaters were tested under maltuned conditions. The pollutant emission rates of the heaters were also determined with the fuel consumption rates adjusted to approximately half their full-input levels. All emission rates are expressed as mass of pollutant emitted per caloric value of fuel consumed.

In addition to the laboratory tests, indoor air pollution levels resulting from the use of four of the eight UVGSHs were tested under various conditions in a partially furnished, unoccupied research house. The fuel input ratings of the heaters used in this phase of the study ranged from 12,600 kJ/h (12,000 Btu/h) to 31,600 kJ/h (30,000 Btu/h). In general, the heaters were operated longer during the field tests than during the laboratory tests.

In this report, we compare laboratory-derived emission rates with those observed at the field research house and present a summary of two more extensive reports: one investigating the pollutant emission rates of UVGSHs under controlled laboratory conditions,  $^6$  and one investigating the indoor air pollution concentrations due to the emissions of UVGSHs under controlled field conditions.'

#### Experimental

Most of the instrumentation used in the laboratory and field experiments was contained in the Lawrence Berkeley Laboratory's Mobile Atmospheric Research Laboratory (MARL). All laboratory measurements were conducted in the environmental chamber, while all field measurements were conducted in the

experimental research house. A single-equation, mass-balance, indoor air quality model was used to calculate pollutant emission rates and also to generate theoretical indoor pollution concentration curves.

#### Laboratory Facilities

Figure 1 shows the schematic of the environmental chamber and supporting instrumentation. The environmental chamber is a  $27-\text{m}^3$  structure housed within a larger building that buffers the chamber from wind, temperature and outdoor pollutant concentration fluctuations. The ventilation rate of the chamber can be varied mechanically from 0.25 to 7.0 air changes per hour (ach). Six miniature "instrument" fans are located inside the chamber to promote air mixing. Each fan has a maximum flow rate of 60  $m<sup>3</sup>/h$  and was located at least one meter from the heater. The axes of the fans were perpendicular to a line between the fan and the heater to avoid air drafts near the heater.

The chamber was cooled primarily by heat transfer through the walls and an interior "cold wail" designed to absorb radiant heat emitted by the heater. The cold wall was composed of two black solar panels that were water cooled.

A nichrome wire heating coil wrapped around glass tubing was placed on the thermocouple of each heater to allow fast starting and to avoid pollutant emissions from the pilot light. A piezoelectric sparker was used to ignite the burners.

#### Field Facilities

Figure 2 shows a schematic of the experimental research house and supporting instrumentation. Again, MARL housed the majority of instruments. The instrumentation used in the field study was similar to that used in the laboratory phase.

All field measurements were made at the unoccupied partially-furnished 107  $m^2$  (1150 ft<sup>2</sup>) one-story research house with an air volume of 240  $m^3$ . Figure 2 illustrates 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 would be well-defined. No mixing fans were used in the house. Doors and windows were slightly opened to achieve air exchange rates representative of typical houses. Air exchange rates were varied by opening doors and windows from 0.5 cm to 3.2 cm creating an<sub>g</sub> additional<sub>g</sub> leakage area in the building envelope of approximately 400  $\texttt{cm}^{\texttt{2}}$  to 1400  $\texttt{cm}^{\texttt{2}}$  and total air exchange rates between  $0.36$  h<sup>-1</sup> and  $1.14$  h<sup>-1</sup>. All interior doors to rooms were open for all field tests.

#### Instrumentation

Schematics of laboratory and field instrumentation are shown in Figures 1 and 2, respectively. The MARL, the primary instrument station, is capable of remote multipoint sampling of  $CO$ ,  $CO_2$ ,  $NO$ ,  $NO_2$ ,  $O_2$ , temperature, dew point, and outdoor wind speed. The MARL sampling system is capable of continuously

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drawing air samples through Teflon tubing from four locations and can use a multipoint timing system to automatically switch between sites at pre-set intervals. Teflon filters are placed at the inlets of the sampling lines to protect the instruments from particulate matter.

During laboratory tests outdoor measurements of all gaseous pollutants except HCHO were made 15 minutes before and after each test. Initial chamber pollutant measurements were also made before the heaters were operated. Outdoor and Indoor HCHO measurements were made simultaneously. During field tests,  $CO_2$ ,  $CO_2$ ,  $NO_2$ ,  $NO_2$ , and  $O_2$  concentrations were recorded every minute. Outdoor, kitchen, living room, and bedroom pollutant concentrations were measured on a rotating basis every six minutes. Data generated during this rotating cycle were analyzed by discarding the first three data points and averaging the last three data points to obtain one observation every six minutes, i.e., twenty-four minutes elapsed between observations at a single site. To eliminate sample-line purge time, air was drawn continuously into the MARL from each site. Temperature and dew point, measured at each of the four locations, and wind speed, measured 6 m above the top of the house, were recorded every minute. A more complete description of the MARL is contained in the report on laboratory investigations.<sup>0</sup>

Formaldehyde levels were measured using a refrigerated bubbler sampler to collect the formaldehyde for subsequent laboratory analysis.  $8$  HCHO sampling times were one hour during laboratory tests and one to two hours for field tests. The concentrations of suspended particles below  $0.5 \mu m$  in diameter were measured using an electrical mobility analyzer.<sup>9</sup> Typically, particulate measurements were taken every 10 minutes.

#### Modeling

A single-equation, mass-balance, model was used to calculate UVGSH pollutant emission rates from measured laboratory data using a technique previously published.  $10$  The model has been used successfully to predict indoor air pollution levels as well as determine indoor air quality parameters that can affect such levels.<sup>5, 10-12</sup> The basic model is repeated here  $\frac{1}{5}$  or  $\frac{1}{10}$  reference; the reader is referred to earlier reports for more information.<sup>7</sup>

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

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

where:

 $C =$  indoor pollutant concentration (ppm);  $C_0$  = outdoor pollutant concentration (ppm); = fraction of the outdoor pollutant level that penetrates the building shell (unitless);  $a = air$  exchange rate in air changes per<sub>3</sub>hour (ach) (h<sup>-1</sup>); S = indoor pollutant source strength  $(cm<sup>3</sup>/h)$ ;  $V = volume(m^3);$  $k$  = net rate of removal process other than air exchange  $(h^{-1})$ .

For particles, C and C<sub>o</sub> are in units of  $\mu$ g/m<sup>3</sup> and S is in units of  $\mu$ g/h. Assuming  $C_{\alpha}$ , P, a, S, and k are constant over the period of interest, Equation (1) can be solved for  $C(t)$  to give:

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

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

Solving Equation 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  $\text{cm}^3/\text{kJ}$  for gases):

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

For gases, E, in  $cm^3/kJ$ , can be converted to  $\mu$ g/kJ by using the ideal gas law and the time-weighted average temperature and pressure in the chamber. The penetration factor, P, was assumed to be 1.0 for gases and 0.4 for parti $cles.$ <sup>10</sup> Note that Equation 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 our measurement of C(T).

#### Laboratory Test Protocol

Dynamic Tests. The experimental protocol for dynamic tests of emission rates was based on the emission rate model parameters listed in Equation 3. The volume (V) of the chamber was determined by physical measurement to be 27 m<sup>3</sup>. The fuel consumption rate, R, was measured using a dry-test gas meter and the appliance combustion time. The heat of combustion of the natural gas was 31.4 kJ/L (1050 Btu/ft<sup>3</sup>), assumed constant during laboratory and field testing. (The local gas utility confirms that the heat of combustion of the supplied natural gas is nearly constant; at worst it varies by only a few percent.) Prior to determining emission rates, all heaters were tuned by adjusting the air shutter for a minimum output of carbon monoxide (as measured by a portable analyzer) and visually observing the flame characteristics. For the partial input tests, fuel consumption rates were set by moving the regulator valve on the heaters to an intermediate setting between "pilot" and "on" to obtain a flame approximately one half its normal height.

After the mixing fans were turned on, the ventilation rate was set, and indoor and outdoor background measurements were made, the heater was ignited and allowed to consume 5540 kJ (5  $ft^3$ ) of natural gas. 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, a, the reactivity constant, k, and the peak concentration, C(T), for each pollutant. Following the decay period, pollutant levels outside the chamber were measured for fifteen minutes.

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As noted earlier, HCHO was measured differently from the other pollutants. Because HCHO can load up in sampling lines, the HCHO sampling lines were periodically purged with nitrogen between tests. The indoor HCHO background was measured periodically and generally agreed with the outside concentrations taken during the tests. A one-hour average measurement of HCHO concentration in the chamber was made during the decay portion of the test and, simultaneously, HCHO was measured outside the chamber.

Steady-state Tests. For steady-state tests, the pollutants were monitored while running the heater in the chamber at a low ventilation rate until the desired  $0<sub>2</sub>$  level was reached. The ventilation rate was then adjusted to maintain a  $\frac{2}{3}$  concentration. In general, monitoring was continued until all pollutants being measured reached equilibrium.

#### Field Test Protocol

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

The UVGSHs were operated in either the living room or the bedroom as shown in Figure 1. Fuel consumption was measured using a dry test meter directly upstream from the heaters. Fuel line pressure was checked and set within manufacturers' specifications. As in the laboratory, precautions were taken to prevent the heater's pilot light from burning until the main burner was lit and a piezoelectric sparker was used to ignite the burner. In each experiment background concentrations were measured indoors for approximately one hour before the UVCSH was turned on and outdoor measurements were made every twenty-four minutes throughout the tests.

Since average concentrations of the whole house are required for the mass-balance model, the gas-phase pollutant data from the three indoor locations were usually averaged in this report. This technique produces an estimate of the whole-house average pollutant concentration every twenty-four 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 the rise of the  $CO<sub>2</sub>$  concentration and the air exchange rate determined during the decay of  $CO<sub>2</sub>$ . The "rise" air exchange rate was determined by using a generalized least-squares computer program to fit the  $CO<sub>2</sub>$  rise data to Eq. (2). For this procedure we set P equal to 1, and k equal to 0, and used the laboratoryderived source strength of CO<sub>2</sub> (51,100  $\mu$ g/kJ x fuel consumption rate). The "decay" air exchange rate was also determined from Eq. (2) using a generalized least-squares technique, but in this case the  $CO<sub>2</sub>$  source strength was zero. The difference between the decay and rise air exchange rates averaged  $0.04 \pm$  $0.10$  h<sup>-1</sup> which is not significantly different from zero.

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#### Results and Discussion

#### Laboratory Results

Eight UVGSHs manufactured by three companies were tested for their consumption of  $O_2$  and emissions of  $CO_2$ ,  $NO_2$ ,  $NO_2$ ,  $HCHO$ , and submicron parti-<br>cles. Each heater was assigned an identification code consisting of two cles. Each heater was assigned an identification code consisting of numbers followed by a letter. The first two numbers, when multiplied by 1000, are the manufacturers' heat input rating in units of Btu/h (1.054 kJ/h) and the letter represents one of the three heater manufacturers, coded as A, B, and C. Tests were made at full input (highest fuel consumption rate) and partial input (approximately one-half the full input rate). Tuning tests and steady-state tests were also conducted on some of the heaters.

Full Input Tests on Well-Tuned Heaters. Table 1 presents the test results for well-tuned heaters operating at full input. "Well tuned" or "good tuning," except when otherwise indicated, denotes the intention of the test rather than an assessment of the results. That is, we attempted to optimize the tuning of a heater by adjusting the air shutter; however, after the test was completed, it was sometimes evident that the heater was not optimally tuned. Except in the case of the 30A heater, no re-adjustments were made since the consumer would not have readjusted the heater. The  $CO<sub>2</sub>$  emission rates (the average of individual tests incorporated in Table 1 is 51,100  $\mu$ g/kJ) and the 0<sub>2</sub> consumption rates (the average of individual tests incorporated in Table 1  $\bar{1}s$  70,900  $\mu$ g/kJ) were relatively constant for all UVGSHs. The relative standard deviation of the measurements for  $CO_2$  and  $O_2$  were both 3%. Based on a spot check of the composition of the natural gas used (Pacific Gas and Electric, San Francisco, CA) we calculated a theoretical emission rate of 51,000  $\mu$ g/kJ for CO<sub>2</sub> and a theoretical consumption rate of 73,200  $\mu$ g/kJ for O<sub>2</sub>, both values are consistent with those measured. However, this agreement should be viewed only as an indicator of accuracy, since the composition of natural gas varies periodically by a few percent and was not routinely checked.

The NO and NO<sub>2</sub> emission rates of the eight heaters averaged 17.3  $\pm$  4.2  $\mu$ g/kJ and 14.1  $\pm$  4.5  $\mu$ g/kJ respectively. The average emission rate for nitrogen of the nitrogen oxides (NO = NO +  $\text{\tt NQ}_2$ ) was 12.6  $\pm$  1.5  $\text{\tt \tt \mu g/kJ,}$  higher than that associated with a gas-fired range.<sup>10</sup> Although the NO<sub>x</sub> emission rates were<br>fairly consistent among all eight well-tuned heaters, the emission rates for NO and  $NO<sub>2</sub>$  in individual heaters showed more variation. Reasons for this variation will be discussed in the section on tuning tests.

As noted, the CO emission rates were much more variable than those of other pollutants. Five heaters had CO emission rates of less than 30 µg/kJ and the other three heaters had much higher rates, up to  $165 \text{ }\mu\text{g/kJ.}$  bther researchers investigating natural gas combustion appliances have observed that the CO emission rates across appliances appear to be log-normally distributed.<sup>13</sup> Our results are consistent with this observation. The geometric mean of the CO emission rate is  $34 \mu g/kJ$ , and the geometric standard deviation is 2.80. Using alternate notation, the geometric mean of the CO emission rates is 34  $(X/\frac{1}{2}$  2.80)  $\mu$ g/kJ.

HCHO emission rates were also assumed to follow a log-normal distribution since, like CO, HCHO is a product of incomplete combustion. The geometric mean emission rate was  $0.81$  (X/ $\div$  2.04)  $\mu$ g/kJ. Heater 12A had the highest HCHO emission rate,  $4.2 \mu g/kJ$ , based upon three tests. This heater was also the only heater to operate below its rated input.

Heater 12A also had the highest particulate emission rate,  $0.32 \mu g/kJ$ , for particles between 0.0056 to 0.56  $\mu$ m in diameter. The particulate emission rates from all UVGSHs followed a log-hormal distribution with a geometric mean of  $0.038$  (X/ $\frac{1}{2}$  3.52)  $\mu$ g/kJ.

In order to assess the reproducibility of the emission rate results, five tests were repeated twice and two tests were repeated three times. The seven sets of tests included experiments with six heaters under well-tuned and maltuned conditions, at low and high ventilation rates and one set was conducted at partial input. A coefficient of variation was computed for each pollutant of the seven sets of repeat experiments. The means of the coefficients of variation for the pollutant emission rates and  $0<sub>2</sub>$  consumption rates were: 35% for CO; 3.0% for CO<sub>2</sub>; 2.6% for O<sub>2</sub>; 14% for NO; 15% for NO<sub>2</sub>; 7.7% for N (of  $NO<sub>x</sub>$ ); 25% for HCHO; and 53% for submicron particles. (Only three data sets were used to determine the mean coefficient of variation for submicron particles due to numerous emission rate values that were below detection.) The range of the coefficients of variation were: 5.3 to 80% for CO;; 0.3 to 5.0% for  $CO_2$ ; 1.0 to 5.3% for  $O_2$ ; 2.2 to 35% for NO; 6.2 to 27% for NO<sub>2</sub>; 2.0 to 16% for N (of NO<sub>v</sub>); 17 to 39% for HCHO; and 6.6 to 83% for submicron particles.

Based on the low variation of the  $CO_2$  and  $O_2$  replicate measurements, the precision of the overall emission rate determination technique used in this report appears to be excellent. It appears that varying emissions from the heaters themselves account for the relatively large variations observed in the CO, NO, NO<sub>2</sub>, N (of NO<sub>x</sub>), HCHO, and submicron particulate emission rate measurements since the observed variation is greater than the precision of their instruments and greater than the coefficients of variation for the  $CO_2$  and  $O_2$ emission rates. The precision of the CO and  $NO/NO<sub>2</sub>$  analyzers are on the order of 1%, as are the precision of the  $CO_2$  and  $O_2$  analyzers. The precision of our HCHO measurement technique is approximately 15%. The high coefficients of variation of the submicron particulate measurements are probably due to varying emissions from the heater and the fact that measurements were made near the limit of detection of the analyzer. The submicron particulate analyzer manual did not specify a precision rating.

Partial Input Tests on Well-Tuned Heaters. As noted earlier, our tests were conducted on UVGSHs operating at full and partial input. Although all three manufacturers contacted insisted that these heaters were not designed to be used at partial input, we found that all heaters tested were capable of being operated at partial input without difficulty or apparent deterioration in performance. The range of input adjustment, while not large, allows the user to obtain a more constant indoor temperature, by eliminating the large temperature fluctuations of on and off operation. Although deemed by the manufacturers to be a misuse of the product, operating at partial input may be common among users.

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Table 2 shows the pollutant emission rates obtained while operating the UVGSH at partial input. The average emission rates for  $CO_2$  and  $O_2$  were 52,100  $\pm$  1,200 µg/kJ and -70,600  $\pm$  3,700 µg/kJ, respectively. These values are very close to the theoretical values mentioned previously. The NO,  $NO_2$ , and N (of NO<sub>v</sub>) average emission rates were 13.0  $\pm$  2.0  $\mu$ g/kJ, 10.2  $\pm$  2.2  $\mu$ g/kJ and 9.2  $\pm$ 1. I µg/kJ, respectively. The geometric means for CO and HCHb emission rates were  $17.2$  (X/ $\div$  2.1)  $\mu$ g/kJ and 0.52 (X/ $\div$  3.3)  $\mu$ g/kJ, respectively. The geometric mean for submicron particulate emissions was determined to be less than or equal to 0.005 pg/kJ. Although not true for each heater, on average the emission rate for each pollutant measured, except  $CO_2$ , was lower during partial input operation than during full input operation.

Tuning Tests. The variability of CO emissions prompted a series of tests on the sensitivity of emission rates to adjustments of the air shutter. All heaters had previously been tuned at full-input with a portable CO analyzer and by visual inspection of the flame characteristics. With adjustment of the air shutter as the only variable, we measured peak<sub>2</sub>CO,  $NO_2$ , and  $NO$  concentrations from heater 30A after combusting 142 L (5 ft<sup>3</sup>) of natural gas under constant ventilation conditions (0.4 ach). The results of these tests are shown in Figure 3 which is similar to an emission versus air/fuel ratio plot with the abscissa representing the percent opening of the air shutter. Throughout the range of shutter settings depicted, 0 to 42% of fully open, the visual flame characteristics are good. The flame characteristics begin to deteriorate only when the shutter is opened beyond 42%. With the air shutter opening increased from 21% to 32% of fully open, the CO concentration increased by a factor of nine. This increase in the air-shutter opening required less than a 10 degree rotation of the shutter, illustrating the sensitivity of CO emissions to tuning.

Although NO<sub>x</sub> emissions are not quite as sensitive to tuning as the CO emissions, the  $NO<sub>2</sub>$ -to-NO<sub>x</sub> ratio is very sensitive. The  $NO<sub>2</sub>$ -to-NO<sub>x</sub> ratio varies from approximately  $\overline{0.2}$  at a 21% shutter opening to almost 1.0 at a 42% shutter opening.

As would be expected, the minimum CO emission occurs near the same shutter setting as the maximum  $NO<sub>v</sub>$  production, i.e. the flame is hottest when there is the least amount of incomplete combustion. However, finding an  $NO<sub>2</sub>$  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<sub>2</sub> was 13.7  $\mu$ g/kJ, HCHO was 1.1 µg/kJ, and submicron particulate emissions were less than 0.004 µg/kJ. With The air shutter fully open the CO emission rate was  $517 \text{ }\mu\text{g/kJ}$ , NO was 0.04  $\mu$ g/kJ, NO<sub>2</sub> was 11.5  $\mu$ g/kJ, HCHO was 20.3  $\mu$ g/kJ and submicron particulate emissions were below 0.004 µg/kJ. The very high CO and HCHO emissions with the shutter fully open aga'in demonstrate how important tuning can be to an appliance pollutant emission rate.

The 40C heater was also tested for pollutant emission rates with its shutter closed and fully open. The results show that the 40C heater was relatively insensitive to tuning compared to the 30A heater. With the 40C heater air shutter closed, the CO emission rate was 35  $\mu$ g/kJ, NO was 13.7  $\mu$ g/kJ, NO<sub>2</sub> was  $11.2 \text{ }\mu\text{g/kJ}$ , HCHO was 0.22  $\mu\text{g/kJ}$ , and the submicron particulate emission

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rate was 0.007 µg/kJ. With the air shutter fully open, the CO emission rate was 8  $\mu$ g/kJ, NO was 19.9  $\mu$ g/kJ, NO<sub>2</sub> was 8.4  $\mu$ g/kJ, HCHO was 0.49  $\mu$ g/kJ, and the submicron particulate emission rate was less than  $0.004 \text{ }\mu\text{g}/\text{kJ}$ . The CO, NO, and HCHO emissions of the 40C heater did not vary as much with shutter setting as did those of the 30A heater. Manufacturer C incorporates a burner design which is very different from the burners of the other two manufacturers. Instead of having many small circular ports in a flat, rectangular burner that produce many small "flamelets," the 40C burner has relatively few slots cut across a cylindrically-shaped burner which produces a softer "feathered" flame. It is likely that this burner design accounts for both the lower emission rates and the insensitivity to air shutter settings.

Steady-state Tests. A series of tests were conducted on the 30A, 16B, and 30C heaters to Investigate steady-state pollutant concentrations at 18%, 19%, and 20% steady-state oxygen levels and at various shutter settings. Table 3 summarizes the results of these tests. (In all the tests reported on Table 3, both  $CO_2$  and  $O_2$  reached steady-state, but because the pollutant emission rates for  $c\ddot{\text{o}}$ , NO, and NO<sub>2</sub> can change with changing O<sub>2</sub> level, they did not always reach steady-state. Those cases are identified in Table 3.)

The last column in Table 3,  $-\Delta$ CO/ $\Delta$ O<sub>2</sub>, gives an index of tuning with respect to CO emissions. These results confirm our earlier observation that the 30A heater emissions are more sensitive to the air shutter settings than are the 16B or 40C heaters. Although the 30A can be well-tuned or maltuned, it appears difficult to maltune the 40C heater and just as difficult to tune the 16B heater.

Since indoor pollutant levels are coupled to steady-state  $0<sub>2</sub>$  levels, the pollutant levels reported in Table 3 may be applicable to residences. The  $NO<sub>2</sub>$ levels measured in residences may be lower than those reported here at the same  $0_2$  level if the NO<sub>2</sub> reactivity rate is greater than 0.3 h<sup>-1</sup>. The NO<sub>2</sub> reactivity rate for our chamber is estimated at  $0.3\,$  h<sup>-1</sup> based on previous chamber experiments.

It is apparent that, in general, higher pollutant concentrations occur at 18%  $0_2$  than at 19% or 20%  $0_2$ . Nevertheless, high pollutant concentration can occur at 20%  $0_2$ . For example, CO reached 90.9 ppm at 20%  $0_2$  using the 30A heater (shutter 52% open). This value is over twice the EPA one-hour outdoor standard for CO of 35 ppm and well above the EPA eight-hour outdoor standard of 9  $ppm.$ <sup>14</sup>

It is interesting to note that all of the CO levels for the 16B heater at 18% and 19%  $0_2$  were well above the EPA one-hour CO standard. All of the CO<sub>2</sub> values measured at 18 and 19%  $0<sub>2</sub>$  for all heaters were above the US occupational eight-hour standard of  $5000$   $ppm<sub>z</sub>$ <sup>15</sup> All of the NO<sub>2</sub> values measured were at legst three times higher than the California one-hour outdoor standard for  $NO<sub>2</sub>$ .  $^{16}$ 

#### Field Test Results

Eighteen controlled field tests were conducted on four UVGSHs. A description of the basic parameters of each test is contained on Table 4. Two of the heaters were used for tests assessing the affects of varying air exchange rates on indoor pollutant levels. The air exchange rates varied from 0.37 to

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l.14 air changes per hour (ach). The average coefficient of variation of the rise and decay air exchange rates was 10%, which gives an estimate of the uncertainty in the air exchange rate values and/or an estimate of the variation in the actual air exchange rates themselves. The indoor/outdoor temperature difference at the end of the UVGSH tests varied from 4.6 <sup>O</sup>C (8.3 <sup>O</sup>F) to 22.6  $^{\circ}$ C (40.7  $^{\circ}$ F).

During the field tests it was observed that the pollutants mixed rapidly throughout the house. This was presumably due to the forced convection driven by the heater. For example, the kitchen, living room, and bedroom  $CO<sub>2</sub>$  concentrations at the end of all UVGSH burns were within 14% of their mean for all tests and, on average, were within 8%.

It is useful to discuss the field results by pollutant since different parameters affect individual pollutants differently. For example, CO emissions are very sensitive to tuning while  $CO<sub>2</sub>$  emissions are not.

Carbon Dioxide and Oxygen. The final concentrations and "projected" steady-state concentrations of  $CO<sub>2</sub>$  and  $O<sub>2</sub>$  for all field tests are listed on Tables 5 and 6. The final concentrations represent the last whole-house measurement made before the UVGSH was turned off. Steady-state conditions were not always reached before the UVGSH was turned off. Therefore, for all tests, the final pollutant concentration was a percentage of the steady-state concentration. That percentage was theoretically determined using the measured house air exchange rate and the combustion time in Eq. (2). 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 short, two-hour experiments (Test  $#30C-1$ ,  $#12B-1$ ).

Graphs of the projected steady-state concentrations for  $CO<sub>2</sub>$  and  $O<sub>2</sub>$  versus the house air exchange rate are shown on Figures 4 and 5, respectively. The curved lines drawn on Figures 4 and 5 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 demonstrate that the steady-state concentrations of nonreactive gases with a constant source strength 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 is known, S can be calculated. For  $CO_2$ , S was calculated to be  $51,600$   $\mu$ g/kJ for the 30A/30C heaters and 53,500  $\mu$ g/kJ for the 16B heater. These values are within 5% of both the laboratory-derived emission rate of  $51,100$   $\mu$ g/kJ and the theoretical emission rate of  $51,000 \text{ µg/kJ.}$ 

The source strengths of  $0<sub>2</sub>$  were analyzed from the data in Figure 5 in a similar manner. The results showed  $0<sub>2</sub>$  consumption rates of 68,900  $\mu$ g/kJ for the 30A/30C heaters and 65,200  $\mu$ g/kJ for the 16A heater. These values are within 9% of the laboratory derived  $0<sub>2</sub>$  consumption rate of 70,900  $\mu$ g/kJ and within 11% of the theoretical  $0<sub>2</sub>$  consumption rate of 73,200  $\mu$ g/kJ.

Although the depletion of  $0<sub>2</sub>$  at the levels observed is not expected to be a health hazard,  $CO<sub>2</sub>$  levels do exceed existing air quality standards and guidelines. The U.S. Occupational Safety and Health Administration (OSHA) has promulgated a time-weighted average  $\textsf{CO}_2$  occupational standard of 5,000 ppm for an eight-hour exposure. The American Society of Heating, Refrigerating, and

Air conditioning Engineers, Inc. (ASHRAE) has recommended a CO<sub>2</sub> "continuous guideline" of 2500 ppm.<sup>17</sup> 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 below  $0.8$  ach. The steady-state  $Co<sub>2</sub>$ levels generated from the 16B heater approached the OSHA standard at 0.46 ac and exceeded the ASHRAE standard for all experiments below 0.9 ach.

Carbon Monoxide. The final observed CO concentrations were below 25 ppm, ranging from 1.0 to 23.2 ppm, with the exception of one test (test 30A-9), which reached 89.4 ppm. Table 7 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  $CO<sub>2</sub>$  and  $O<sub>2</sub>$  and are shown graphically on Figure 6.

As shown in our laboratory studies, carbon monoxide emission rates are not constant from heater-to-heater nor are they constant for an individual heater if the air/fuel ratio changes. These facts necessitated a different analysis strategy for CO than 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 under 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, 5, of CO was allowed to vary while the air exchange rate was constrained to be the value determined using  $CO<sub>2</sub>$  and listed on Table 4. It was necessary to constrain the air exchange rate since 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 actual initial concentration, our procedure does not give as much statistical weight to the early points as the later points. A list of these regression-derived "average" emission rates are included in Table 7.

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 adjacent whole-house CO and CO<sub>2</sub> estimates were inserted into Equation (3) as  $C(0)$  and  $C(T)$ . (The parameters in Equation (3) were determined elsewhere.) This step yielded "uncorrected" CO and  $CO_2$  "semi-continuous" 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 semi-continuous CO emission rate by reducing the effect of incomplete mixing, which should affect  $CO<sub>2</sub>$  concentrations in the same manner as it affects CO.

The results of this semi-continuous source strength analysis shows that CO emissions can change with time. This is demonstrated in Table 7 which shows that the initial emission rate of CO, determined within 5 to 15 minutes of ignition, can be very different than the regression-derived "average" emission rate or the final emission rate, defined as the average of the last three semi-continuous emission rate points within the last 72 to 85 minutes of the burn. In general, the CO emission rate profiles were not monotonic and allow

the possibility that the average CO emission rate is not between the initial and final values.

The regression-derived emission rates of the 30A heater under well-tuned conditions ranged from 2.4 to 16  $\mu$ g/kJ, lower than the 25  $\mu$ g/kJ measured in our laboratory tests. However, the average of the initial CO' emission rates was 25  $\pm$  6 µg/kJ which agrees very well with our laboratory test results which had a combustion time of approximately ten minutes.

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

The regression-derived CO emission rate and initial CO emission of the 30A under the excess air condition (shutter 69% open) of 620  $\mu$ g/kJ and 780  $\mu$ g/kJ respectively, were above the laboratory-derived emission rate of 517  $\mu$ g/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 settings greater than 69% open.

The CO emission rates from the 16B heater were very constant over time, especially when compared to the other heaters. The laboratory emission rate of 190 µg/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, 16B-2). The mean of the regression-derived emission rates for tests  $16B-3$  through  $16B-7$  was  $200 \pm 10$  $\mu$ g/kJ and the mean of the initial emission rate values was 200  $\pm$  30  $\mu$ g/kJ which shows the aforementioned agreement.

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

Carbon monoxide emission rates can vary substantially from heater-toheater, condition-to-condition, test-to-test, and even with time. The reasons for the variation in CO emission rates are not well understood, but it is apparent that tuning (or, more correctly, the air/fuel ratio) plays a major role in the variations observed within a single heater. It is also apparent that the CO levels in a house with an UVGSH can exceed outdoor long-term and short-term standards. Nine of the eighteen 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 using the 30A heater under the excess air condition.

Nitrogen Oxides. The test results and calculations for NO,  $NO_2$ , and  $NO_X$ are reported on Tables 8, 9, and 10, respectively. Actual levels observed ranged from  $0.00$  to 5.14 ppm for NO; 0.225 to 1.35 ppm for NO<sub>2</sub>; and 1.21 to 6.36 ppm for  $NO<sub>x</sub>$ . To compare the consistency of the pollutant levels observed

in the field with those based on laboratory tests, it was necessary to derive "average" field emission rates and semi-continuous emission rates for NO,  $NO<sub>2</sub>$ , and N (of  $NO_x$ ). This was accomplished in a similar manner as reported with  $CO$ with the exception that a reactivity term was needed for NO, NO<sub>2</sub>, and NO<sub>x</sub>. 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.  $CO_2$ , 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. Measured reactivities are listed on Tables 8-10.

The average reactivity rates for NO and NO<sub>x</sub>,  $0.04 \pm 0.08$  h<sup>-1</sup> and  $_{\rm c}0.08 \pm 0.08$  $0.08$   $\rm h^{-1}$  respectively, are similar to those observed by Traynor et al.<sup>5</sup> in the same house using a gas-fired range as the pollutant source, but the average  $NO<sub>2</sub>$  reactivities were much lower in these tests than those reported earlier, The NO<sub>2</sub> reactivities of the tests in this report <sub>l</sub>averaged 0.20  $\pm$  0.13 h<sup>-</sup> while the earlier study reported 1.29  $\pm$  0.67 h<sup>-1</sup>. 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, indicating that water vapor or the  $NO<sub>2</sub>$  deposition rate on painted surfaces may play a role in the overall NO<sub>2</sub> reactivity rate. However, there were no significant differences in the relative humidity levels between the two studies.

Using the calculated reactivity rates along with the air exchange rate it. was possible to determine how close our final whole-house concentration measurement was to steady-state. The NO<sub>2</sub> levels for the 30A and 16B heater tests reached between 83.4% and 99.9% of steady-state (see Table 9). Projected steady-state levels were calculated and are shown on Figure 7.

The mean of the regression-derived emission rates for N (of NO<sub>y</sub>) from the 30A heater under well-tuned conditions was  $15.7 \pm 2.2$  µg/kJ which is slightly above the laboratory rate of 13.6  $\mu$ g/kJ. The initial emission rates for the same tests averaged 13.7  $\pm$  1.4  $\mu$ g/kJ, a value much closer to the laboratoryderived rate. The mean of the regression-derived  $NO<sub>2</sub>$  emission rates for these tests was  $7.5 \pm 1.9 \text{ µg/kJ}$  which was lower than the laboratory rate of 11.4  $\mu$ g/kJ. However, the initial NO<sub>2</sub> emission rate, averaging 10.4 ± 3.0  $\mu$ g/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 N (of NO<sub>v</sub>) emission rate,  $11.1 \pm 0.3$  µg/kJ, is closer to the laboratory rate of 11.2 µg/kJ than is the  $\,$  mean of the <code>'regression-derived</code> emission rates,  $12.8\,\pm\,0.7$   $\mu{\rm g}/{\rm kJ}$ . And the average initial NO $_{\gamma}$  emission rate of  $11.6$   $\pm$  2.2  $\mu$ g/kJ is closer to the laboratory rate of  $13.7 \text{ }\mu\text{g/kJ}$  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 we observed no increase in NO levels and thus had an emission rate below 0.1  $\mu$ g/kJ for NO which agrees with the laboratory-derived emission rate of  $0.04 \mu g/kJ$  when the heater air shutter was 100% open. In addition, the initial  $NO_2^{\dagger}$  and N (of  $NO_x$ ) emission rates of 3.0  $\mu$ g/kJ and 9.7  $\mu$ g/kJ, respectively, agreed within 20% of the laboratory rates of 3.4  $\mu$ g/kJ for NO $_2$  and 11.5  $\mu$ g/kJ for N (of NO $_\mathrm{\textbf{x}}$ ). The <code>NO</code> $_2$  and N (of NO $_\mathrm{\textbf{x}}$ ) "average" and final emission rates were approximately twice

#### the initial value.

As noted previously with CO emission, the  $NO_x$  emission rate from the 16B heater under well-tuned conditions did not change drastically with time. The average initial NO,  $NO_2$ , and N (of NO<sub>x</sub>) emission rates for tests 16B-1 through 16B-7 were 16.3  $\pm$  3.6  $\mu$ g/kJ, 18.3  $\pm$  2.8  $\mu$ g/kJ and 13.2  $\pm$  2.4  $\mu$ g/kJ, respectively, and agreed with the laboratory-derived rates of 13.9  $\mu$ g/kJ, 18.1  $\mu$ g/kJ and 12.3  $\mu$ g/kJ for NO, NO<sub>2</sub>, and NO<sub>x</sub>, respectively. The regression-derived NO emission rates averaged  $20-4 \pm 3.4 \text{ }\mu\text{g/kJ}$ , approximately 25% higher than the initial emission rate. The regression-derived  $NO<sub>2</sub>$  emission rates averaged 17.8  $\pm$  1.9  $\mu$ g/kJ, about the same as the initial values.

Results from the 30C and 12A heater tests showed elevated initial  $NO<sub>2</sub>$ emission rates and initial low *NO* emission rates compared to the "average rates. The laboratory-derived emission rates for  $NO_{\chi}$  and  $NO_{\gamma}$  emitted from the 30C heater were 19.3  $\mu$ g/kJ and 9.6  $\mu$ g/kJ, respectively, below initial NO and NO<sub>2</sub> emission rates of 24.1  $\mu$ g/kJ<sup>'</sup> and 12.0  $\mu$ g/kJ, respectively. The laboratory-derived emission rates from the 12A heater were 9.6 µg/kJ for NO and 19.7  $\mu$ g/kJ for NO<sub>2</sub>, which compared favorably to the initial field NO<sub>2</sub> emission rate values  $\bar{o}$ f 8.4 µg/kJ for NO and 19.0 µg/kJ for NO<sub>2</sub>.

It appears that the NO,  $NO_2$ , and  $NO_x$  emission rates can change over time. The  $NO_x$  emission rates do not change as much as  $NO$  and  $NO_2$  emission rates, while NO and NO<sub>2</sub> emissions are, in general, not as variable as CO emission rates. As with  $CO$ , the NO, NO<sub>2</sub>, and NO<sub>y</sub> emission rate profiles are not always monotonic and can lead to initial and final emission rates that are both below the regression-derived rate.

Of NO,  $NO_2$ , and  $NO_x$ , only  $NO_2$  is regulated by outdoor standards. The only outdoor short-term NO<sub>2</sub> standard promulgated by a state or federal agency is the California short-term standard set at  $0.25$  ppm for a one-hour average.<sup>16</sup> The projected NO<sub>2</sub> steady-state concentrations exceeded the California one-hour standard for every test (see Figure 7). All but one of the actual final  $NO<sub>2</sub>$ concentrations also exceeded the California short-term, NO<sub>2</sub> standard. The lone exception was for test  $12A-1$  which consumed only  $9,510^2$  kJ/h and was operated for only two hours. The final  $NO<sub>2</sub>$  level reached in test  $12A-1$  was 0.23 ppm, just under the California short-term standard.

Particulates and Formaldehyde. In general the particulate and formaldehyde concentrations were low. The highest sustained submicron particulate concentration was approximately 4  $\mu$ g/m $^{\texttt{J}}$  during test  $\mathfrak{Z}0$ A-1. This value is well below the long-term EPA outdoor s'tandard of 75 µg/m' for total suspended particulates. 14

Of all the tests with formaldehyde measurements, only one (test 30A-9) had formaldehyde levels above the most stringent of formaldehyde standards,  $0.10$ ppm.<sup>18</sup> A concentration of 1.3 ppm was reached during that test. This was also the test with high CO levels and the only tests conducted with a heater under excess air conditions. An estimate of the HCHO emission rate was made by assuming a reactivity rate of  $0.4 \text{ h}^{-1}$ , based on earlier chamber studies.<sup>10</sup> The field HCHO emission rate was 14  $\mu$ g/kJ which is lower than the 20.3  $\mu$ g/kJ measured in the laboratory with the shutter 100% open. There was 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 consistent.

Additional Discussion. One of the more interesting observations made in this study is the very rapid mixing observed in all tests. This was even true of the 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 easily explained by the thermal storage in the walls, and other interior surfaces which buffer changes in temperature.

Because the air in the house mixes rapidly, the implied assumption in our model of well-mixed air is a good one. On the other hand, our assumption that the source strength and emission rates from UVGSH are constant is not always met. When the constant source strength assumption 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 laboratoryderived rates. The constant source strength assumption is a good one for  $CO_2$ ,  $0_2$ , NO<sub>x</sub>, and, for some experiments, CO. In general, it is not a good assumption for NO and NO<sub>2</sub> individually or for many CO experiments. However, the regression-derived emission rates yield average emission rates over an extended period of time and may be more useful than initial or final emission rates.

#### Conclusions

The three basic goals of this study were to quantify pollutant emission rates in the laboratory, to determine if laboratory-derived emission rates were applicable to a field situation, and to determine actual indoor pollutant levels due to the use of an unvented gas-fired space heater in a real house. The first goal was accomplished and laboratory-derived emission rates were reported. The results concerning the second goal show that laboratory emission rates are, in general, applicable to initial field emission rates, i.e., those within approximately 15 minutes of ignition. They are also applicable in situations when the emission rates are constant over time such as for  $CO_2$ ,  $O_2$ ,  $NO<sub>x</sub>$ , and, under certain conditions NO,  $NO<sub>2</sub>$ , and CO. For NO,  $NO<sub>2</sub>$ , and CO the actual use pattern of the UVGSH is critical in determining indoor pollutant levels. This study also confirmed the importance of tuning in the emission rates. This holds especially true for  $CO$ ,  $NO<sub>2</sub>$ , and HCHO.

With regard to the third goal, we observed levels of  $CO<sub>2</sub>$  and  $NO<sub>2</sub>$  at or above existing standards or guidelines during most steady-state laboratory and controlled field tests.  $\text{NO}_2$  levels were consistently higher than California l-hour outdoor NO<sub>2</sub> standard (O.25 ppm).  $10^{\circ}$  CO levels were often observed over the EPA 8-hour standard (9 ppm)<sup>14</sup> under well-tuned conditions and were sometimes observed to be over the EPA 1-hour standard (35 ppm)<sup>14</sup> under maltuned conditions.  $CO_2$  levels were above the OSHA occupational 8-hour standard (5000 ppm)<sup>15</sup> when the 31,600 kJ/h (30,000 Btu/h) heaters were operated with a ventilation rate below 0.8 ach. HCHO levels greatly exceeded the American Industrial Hygiene Association's guideline  $(0.10 \text{ ppm})^{18}$  during one maltuned test. The lowest projected steady-state  $0<sub>2</sub>$  level in the field tests was 19.08% and all measured levels were above 19.4%.

We must therefore conclude that UVCSHs may pose a potential threat to the health of the occupants of houses where such an appliance is used based on  $NO<sub>2</sub>$ findings alone. Depending upon 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  $CO_2$ ,  $CO_2$ , and HCHO may also be in excess of current recognized health standards and may pose an additional health hazard.

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Table 1. Pollutant emission rates from eight well-tuned, unvented gas-fired space heaters operated at full input<br>In an environmental chamber with low ventilation.



 $a_{\texttt{Identif1cat1on code: } 40B = 40,000 \text{ Btu/h (42,200 kJ/h) here from manufacturer B. Heat content of natural gas used was 31.4 kJ/l.}$ b<sub>percent</sub> of fully open.

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Chass 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<sup>3</sup>. duater subjected to extensive tuning and considered optimally tuned.

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Pollutant emission rates from eight well-tuned, unvented gas-fired space heaters operated at partial input<br>in an environmental chamber with low ventilation. Table 2.

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aldentification code: 40B = 40,000 Btu/h (42,200 kJ/h) heater from manufacturer B. Heat content of natural gas used was 31.4 kJ/L. b<sub>percent</sub> of fully open.

Chass 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<sup>3</sup>. dueater subjected to extensive tuning and considered optimally tuned at full input.

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# Table 3. Pollutant concentration increases above background (outdoor)<br>levels from unvented gas-fired space heaters operating near steady state<br>in a  $27-\pi^3$  chamber.

apid not attain steady-state. Concentration given is the value measured closest to projected steady-state value.

bExceptionally high background values for CO (3.5 to 4.6 ppm) relative to chamber CO value.



Table 4. Description of Controlled Field Tests.

 $a$ Heat content of natural gas used = 31.4 kJ/L. bAverage of indoor kitchen, living room, and bedroom values minus the outdoor value; computed at end of burn.



Table 5. Controlled Field Test Results and Calculations for Carbon Dioxide.

aTime from start of burn to last whole-house average concentration during burn.

 $<sup>b</sup>$  Last whole-house average concentration. Average background CO<sub>2</sub></sup> concentration for all tests is  $320 \pm 30$  ppm.

 $c(1 - e^{-at})$  X 100 where a is air exchange rate  $(h^{-1})$ , t is elapsed burn time  $(h)$ .



Table 6. Controlled Field Test Results and Calculations for Oxygen.

aTime from start of burn to last whole-house average concentration during burn.

 $<sup>b</sup>$  Last whole-house average concentration. Average background  $0<sub>2</sub>$ </sup> concentrations for all tests is 20.92% ± 0.05%.  $c_{(1 - e^{-at})}$  x 100.

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Table 7. Controlled Field Test Results and Calculations for Carbon Monoxide.



atime from start of burn to last whole-house average concentration.

 $b$  ast whole-house average concentration. Average background CO concentration is 0.40 ± 0.24 ppm.  $c_{(1-e}$ -at) X 100.

dpetermined by generalized least squares.

erirst emission rate value within 5 to 15 minutes of ignition.

frime-weighted average of three emission rate values within 72 to 85 minutes of the end of the burn.

Table 8. Controlled Field Test Results and Calculations for Nitric Oxide.

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 $^{a}$ Nean = 0.04 ± 0.08 h<sup>-1</sup>.

brime from start of burn to last whole-house average concentration.

 $c_{\text{Average background concentration}}$  oncentration = 0.008 ± 0.007 ppm.

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

enetermined by generalized least squares.

frirst emission rate value within 5 to 15 minutes of ignition.

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

Table 9. Controlled Field Test Results and Calculations for Nitrogen Dioxide.



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

brime from start of burn to last whole-house average concentration.

 $c_{Average\ background\ concentration} = 0.031 \pm 0.009$ .

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

epetermined by generalized least squares.

frirst emission rate value within 5 to 15 minutes of ignition.

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

Table 10. Controlled Field Test Results and Calculations for Nitrogen Oxides.

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Final<br>  $N$  (of  $NQ_x$ )<br>
emission<br>
rate<sup>8</sup><br>
rate<sup>8</sup><br>
(µg/kJ)  $13.9$ <br> $12.5$  $\begin{array}{c} 13.3 \\ 17.3 \end{array}$  $11.4$ 11.9 11.9 12.9 14.6 17.6 14.7  $\ddot{ }$ .8  $\ddot{\cdot}$ 13.7  $13.2$  $\begin{array}{c} 1 \end{array}$ 11.7 15.1 Initial<br>N (of NO<sub>X</sub>)<br>emission<br>rate<br>rate<br>(µg/kJ)  $\frac{1}{11}$ <br> $\frac{3}{11}$  $\frac{1}{10}$ .0 11461<br>11111 14.9  $13.5$  $3.0$  $9.7$ "Average"<br>N (of NO<sub>X</sub>)<br>emission<br>rate<sup>e</sup><br>rate<sup>e</sup><br>(Hg/kJ)  $\frac{17.0}{15.1}$  $14.3$ <br> $13.3$ <br> $18.7$  $15.0$ <br> $13.2$ 13.9  $11.4$ <br> $11.4$ <br> $12.8$ 16.8  $15.3$  $17.5$ 15.8  $14.2$  $\ddot{6}$ Projected<br>ateady-state  $\sum_{(p) \in \mathbb{N}}$  $7.50$ <br>6.65  $5.23$ <br> $4.30$ <br> $3.48$  $\frac{1}{3}$ .56 1.36  $2.32$  $2.65$ <br> $2.10$ 7.76 2.39 4.83  $3.03$ <br> $2.97$  $1.47$  $3.17$ steady-state<sup>d</sup> Theoretical  $70f$ 19149<br>20030 91.5<br>98.8 98.9  $92.2$ <br>95.3 49.8 94.9 99.8 54.0 94.4 96.4 Pinal<sup>C</sup> **Label** 384024  $4.42$ <br> $4.06$ <br> $3.54$  $1.35$  $2.93$ <br> $2.21$  $2.88$  $2.56$ <br> $2.04$  $1.47$  $1.21$  $4.21$ concentration<sup>b</sup><br>concentration<sup>b</sup> Elapsed<br>time to final 273<br>2374<br>2774 2588251 274<br>388<br>370  $273$ 274 106 106 Reactivity,<sup>a</sup>  $\sum_{i=1}^{k-1}$  $\begin{array}{c} 8 & 2 & 5 \\ 6 & 6 & 6 \\ 9 & 9 & 9 \end{array}$  $0.09$  $\overline{0.11}$  $0.13$  $0.10$ <br> $0.03$  $0.24$  $0.18$ <br> $0.02$  $0.12$  $0.08$  $0.00$  $0.22$ Air<br>exchange  $\int_{\frac{1}{2}}^{\frac{1}{2}}$  $0.57$ <br>0.69  $0.49$ 1.08  $0.45$ <br>0.57  $0.70$  $0.76$  $0.46$ <br>0.64 0.79 0.39  $0.37$  $0.52$  $0.47$ 0.61  $0.36$  $30A-5$  $30A-9$  $30A-3$  $30A - 4$  $30A-6$  $30A - 8$  $16B - 2$  $30A - 2$  $30A-7$  $16B-3$  $168 - 4$  $168 - 5$  $168 - 6$  $30C-1$  $12A-1$ code  $30A-1$  $16B-1$ Test  $16B-7$ 

 $^{a}$  Hean = 0.08 ± 0.08 h<sup>-1</sup>.

brime from start of burn to last whole-house average concentration.

 $c_{\text{Average}}$  background concentration = 0.019  $\pm$  0.014.

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

epetermined by generalized least squares.

Fritst emission rate value within 5 to 15 minutes of ignition.

Brime-weighted average of three emission rate values within 72 to 85 minutes of the end of the burn.







XBL 8110-1378A

Figure 1. Schematic of the environmental chamber and the Mobile Atmospheric Research Laboratory (MARL) used for laboratory experiments.



Figure 2. Schematic of the field research house and supporting instrumentation used for field experiments.



Peak CO,  $NO_2$ , and  $NO_x$  concentrations versus the primary<br>air-shutter opening for 31,700 kJ/h unvented gas-fired, space Figure 3. heater (heater code: 30A) after combusting  $142$  L (5 ft<sup>3</sup>) of natural gas in a  $27-\text{m}^3$  environmental chamber. The chamber air was well-mixed and the ventilation rate was approximately 0.4 air changes per hour.





Figure 4. Projected steady-state  $CO<sub>2</sub>$  concentrations due to the use of unvented gas-fired space heaters versus the house air exchange rate. Tests were conducted in a  $240 \text{ m}^3$  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.





Projected steady-state  $0<sub>2</sub>$  concentrations due to the use of unvented gas-fired space heaters versus the house air exchange rate. Tests were conducted in a 240  $m<sup>3</sup>$ 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 6. Projected steady-state CO concentrations due to the use of unvented gas-fired space heaters versus the house air exchange rate. due to the use of unvented gas-fired space<br>heaters. Tests were conducted in a 240  $\pi^3$  uncompied Tests were conducted in a  $240 \text{ m}^3$  unoccupied research house.



Figure 7. Projected steady-state  $NO<sub>2</sub>$  concentrations due to the use of unvented gas-fired space heaters versus the house air exchange rate. Tests were conducted in a 240 m<sup>3</sup> unoccupied research house.

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