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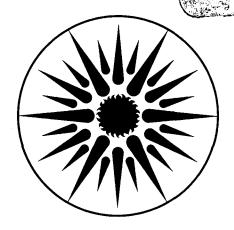
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<u>Abstract</u>

Wood-burning stoves can emit pollution indoors during starting, stoking, and reloading operations. They can also emit pollution indoors if they are not air-tight or are malfunctioning. Four woodburning stoves, three air-tight and one non-air-tight, were operated in a single-floor 236-m³ residence and tested for pollutant emissions. Results showed the air-tight stoves emitted small "puffs" of carbon monoxide and respirable suspended particles during door-opening operations while the non-air-tight stove continuously injected pollutants indoors under certain operating conditions. During the non-air-tight stove operation, carbon monoxide levels reached a maximum of 43 ppm while average suspended particulate concentrations ranged from a typical outdoor concentration of 30 $\mu g/m^3$ up to 800 $\mu g/m^3$. Five polycyclic aromatic hydrocarbons, including benzo(a)pyrene, were measured in the collected particulate samples and the results are Source strengths for carbon monoxide, total suspended reported. particles, and five polycyclic aromatic hydrocarbons are reported for each stove type.

Introduction

Two areas of concern to both the scientific community and the general public are energy conservation and indoor air pollution. Strategies designed to reduce energy costs are not always compatible with acceptable indoor air quality. For example, the increased use of alternative fuels aimed at reducing residential heating cost--such as the shift to wood-burning stoves--can have a detrimental effect on indoor air quality if the alternative-fuel appliance emits pollutants indoors.

Wood-burning stoves have been found, under some conditions, to contribute to indoor concentrations of carbon monoxide, nitric oxide, nitrogen dioxide, and suspended particles containing polynuclear aromatic hydrocarbons (PAH) including benzo(a)pyrene (1-4). Indoor pollutants emitted from wood-burning stoves can enter into the indoor environment during starting and stoking operations, or they can be emitted continuously if a leak or crack exists in the stove or its vent system. In this study we investigated the impact on indoor air quality from the use of four different wood-burning stoves in an occupied house under simulated use conditions. Measurements of carbon monoxide (CO), nitric oxide (NO), nitrogen dioxide (NO2), formaldehyde (HCHO), total suspended particles (TSP), submicron suspended particles (<0.6 μ m) and five PAH compounds are discussed in this paper. The PAH compounds investigated were benzo(b)fluoranthene (BbF),

benzo(k)fluoranthene (BkF), benzo(a)pyrene (BaP), benzo(ghi)perylene (BghiP), and indeno(1,2,3-cd)pyrene (IcdP).

Experimental Methods

Test House

The house used in this study was in Truckee, California (elevation: 1800 m) where wood stoves are common home heating appliances. The house was a single-floor, ranch-type structure with a volume of 236 m³. The house was occupied by the project staff during the tests. Figure 1 shows the floor plan and the location of the stove and air sampling sites. All windows, closet doors, and outside doors were closed during the tests while all bedroom doors were left open.

Wood-burning Stoves

Four models of wood-burning stoves were tested to determine their contribution to indoor air pollution. Basic information on each stove is listed on Table I. Three models were considered air-tight while one was not. The flue vent system was 20 cm (8 inches) in diameter and was equipped with a damper. Stoves A, C, and D were designed for 20 cm flues. Stove B was designed for a 15 cm flue and an adaptive collar was used. Stove A was operated both with its doors opened, as a fireplace, and with its doors closed; stove D was operated with its

doors closed. All stoves had upstream dampers while stove D had both an upstream and a downstream damper.

Instrumentation and Analytical Methods

Gaseous pollutant concentrations, except for HCHO, were measured using the Lawrence Berkeley Laboratory Mobile Atmospheric Research Laboratory (MARL) (5-6). These pollutants were monitored in the living room, kitchen, master bedroom, and outside on a 24-minute rotating cycle, six minutes at each location. HCHO was measured at all four locations using an integrating bubbler/collection system (7). Temperature measurements were made continuously at all four locations. Air exchange rates were measured using an automated sulfur hexafluoride tracer system.

Integrated TSP concentrations were gravimetrically determined using samples collected on Teflon filters. Face velocites across the filters ranged from 70 to 80 cm/sec. Typically, a single filter was used per test at one indoor and one outdoor location. If a filter became heavily loaded, based on an excessive increase in pressure drop across the filter, the filter was changed.

PAH concentrations were determined from the TSP filter samples. One section (2.00 cm²) of the teflon filter was extracted in 0.40 ml peroxide-free spectral quality tetrahydrofuran by sonication at 25°C

for 15 minutes (8). A 50 μ l aliquot of the extract was injected onto a liquid chromatographic column, which had been chosen for its selectivity in separation of PAH compounds (VYDAC 201 TP) (9). A mixture of acetonitrile and tetrahydrofuran (19:1, v:v) in water was used as the mobile phase to separate the sample components. Composition of the acetonitrile-tetrahydrofuran mixture was varied from 70-98% by gradient elution. The separated PAH compounds were detected using fluorescence with an excitation wavelength of 280 nm. All fluorescence \geq 389 nm was recorded as the sample components eluted from the column. Peak heights were used to determine concentrations of PAH in the extracts.

Real-time submicron (< 0.6 μ m) particulate concentrations were monitored with an electrical mobility analyzer (10). Because of the assumptions of spherical shape and 2.0 g/cm³ particulate density used in calculating submicron mass concentrations, their measured absolute concentrations are not as accurate as the TSP concentration measurements. A comparison of TSP data and submicron particulate data shows that the latter (expressed in terms of mass) may be high by a factor of approximately two. The authors recommend that the submicron particulate data be used for inter-test comparisons and not for their absolute concentrations.

Pollutant source strengths were calculated for the four wood stoves tested. Assumptions of a well-mixed house and steady-state

conditions were employed. The steady-state equation used to calculate the source strengths follows.

$$S = [C(a + k) - PaC_0]V$$
 (1)

where: S = pollutant source strengths (mass/h or volume/h);

c = average indoor pollutant concentration (mass/m³ or volume/m³);

a = building air exchange rate (h-1);

k = net rate of removal processes other than air flow (h-1):

P = fraction of outdoor contaminants that penetrate the building shell (unitless)($0 \le P \le 1$);

 C_0 = average outdoor pollutant concentration (mass/m³) or volume/m³);

V = building volume (m³).

The approach used to determine values of a, k, and P has been previously published along with a more detailed discussion of the indoor air pollution model (11). Of interest in this paper is the penetration factor, P, applied to particles of outdoor origin. Using the following equation, the penetration factor can be calculated with data from tests when no wood-burning stove was operated.

$$C(a + k)$$

 $P = ---- Co^{a}$
(2)

Testing Protocol

All tests were designed to simulate actual use conditions. The stoves were initially started using pine kindling. Depending on the test, either pine or oak logs were used to fuel the stoves. Stove fuel consumption was determined by weighing the wood before combustion and subtracting the mass of the charred remains. The wood-burning stoves were operated to keep the indoor air temperature in the range of 20 to 25°C. (Average outdoor temperatures ranged from -2°C to 3°C.) The stoves were operated for a minimum of five hours during each test. A baseline for indoor suspended particles was determined during two days of measurements without a wood-burning stove operating in the house.

Results and Discussion

A summary of combustion times, consumed fuel mass, air exchange rates, CO data, and submicron particulate data is shown in Table II. Table III summarizes the TSP and PAH results. In general, the three air-tight stoves (A, B, and C) emitted only small amounts of pollutants indoors. However, the one non-air-tight stove (D) emitted large amounts of CO and PAH-containing suspended particles.

All air-tight stoves caused_at_least one short-term increase in CO and submicron particulate levels. Figure 2 shows the real-time

pollutant concentration and temperature profile while operating stove A. The initial peak observed in the particulate concentrations corresponds with the starting of the stove. The subsequent peaks were a result of stoking and reloading the stove. The high ${\rm CO_2}$ peak was a result of researchers being inside the house during the initial burn period. These peaks resulted in average CO levels above outdoor levels, however, with the exception of two tests, they were not sufficient to elevate indoor PAH concentrations above corresponding outdoor concentrations. The two exceptions were with the operation of stove A.

The non-air-tight stove, stove D, was a moderate to heavy indoor pollution source depending upon how the stove was operated. Unlike the air-tight stoves, the emissions from the non-air-tight model were very sensitive to how the stove was operated. One reason for this sensitivity was that stove D, a 30-inch Franklin-type model, had visible gaps in the stove's construction which negated the usefulness of the stove's upstream damper for controlling the fire. It was necessary to use the downstream damper to control the fire which reduced the pressure difference between the inside of the wood stove and the house living space. During tests D-2 and D-3 the stove overheated, possibly due to over filling the stove with wood, and it was necessary to reduce the air flow to the fire using the downstream damper. This caused very high indoor levels of CO and submicron particles (see Figure 3). In contrast, the wood level was kept very

low during tests D-1 and D-4 and the resulting CO and submicron particle levels were lower (see Figure 4), but still not as low as the indoor pollutant levels from the air-tight models. (Note the similarity of pollutant concentration levels at the three sampling locations of Figures 2, 3 and 4.) During tests D-2 and D-3 the researchers noticed a large amount smoke indoors which presumably would have been detected by the occupants, thus these two tests should be considered "worst case" tests. (For reference, the U.S. Environmental Protection Agency's outdoor 24-hour TSP standard is $260 \, \mu g/m^3$, the 8-hour CO standard is 9 ppm and the 1-hour CO standard is $35 \, \text{ppm}$ (12); however, the indoor environment was not a consideration in the development of these standards.)

Indoor and outdoor NO and NO $_2$ concentrations were low for all tests conducted. Indoor and outdoor average NO and NO $_2$ concentrations were below 0.02 ppm, except for NO in tests D-2 and D-3, where the average increase in NO concentration was 0.021 ppm and 0.070 ppm, respectively. HCHO concentrations were also low in general. The only stove to cause an increase in HCHO concentrations above the house's normal background was stove D. The HCHO increases ranged from 10 ppb for test D-2 to 49 ppb for test D-3.

The ratio of the sum of the five PAH concentrations to the TSP concentration varied between test types (see Table III). Qualitatively, the PAH/TSP ratio increased with the amount of TSP

originating from wood-burning stoves; however, other factors such as burn rates can affect the PAH/TSP ratio. Note that tests A-2 and D-3 had unusually high ratios for their respective groups and were also tests with relatively uncontrolled air flows across the wood.

Carbon monoxide, TSP and BaP source strengths were calculated using Eq. 1 for all tests and are shown on Table IV. For tests with stove D, an estimate was made for the average TSP and BaP concentrations by utilizing the real-time profiles from the sub-micron particulate analyzer (see Table III). Although many assumptions are implied by using such a technique, it did appear to remove the bias introduced by sampling suspended particles during short time periods that may or may not have been representative of the average TSP and BaP concentrations over the entire test.

In order to calculate pollutant source strength several parameters needed to be determined. The indoor particulate decay rate, k, was calculated to be $0.08\pm0.04~h^{-1}$ using the simultaneous submicron particulate and CO decay data. The average penetration factor for particles of outdoor origin was calculated to be 0.48 ± 0.28 using Eq. 2 and all the PAH data from the two control tests. This average value is consistent with those observed by other researchers (13-15). The penetration factors we observed, using individual PAH compounds, fell into two groups. The penetration factor for BbF, BkF and BaP averaged 0.28 ± 0.03 while the factor for

BghiP and IcdP averaged 0.78 ± 0.12 . Possible reasons for these different apparent penetration factors will be discussed later.

The source strength data on Table IV illustrate many points. First, even without a wood stove operating there appeared to be a source of TSP. Since the house had no other obvious source of suspended particles, we conclude that house "dust" was suspended or resuspended by the researchers in the house or by some other unidentified mechanism. Second, we again observe the great difference between the emissions from the air-tight versus non-air-tight models. Finally, we observe the wide variation in emission rates for the non-air-tight stove, a result of different user-controlled operating styles.

Source strengths for BbF, BkF, BaP, BghiP, and IcdP averaged 0.03 \pm 0.02 μ g/h, 0.01 \pm 0.01 μ g/h, 0.05 \pm 0.03 μ g/h, 0.03 \pm 0.03 μ g/h, and 0.06 \pm 0.05 μ g/h, respectively, for air-tight stove tests, excluding test A-2. When stove A was opeated as a fireplace (test A-2), the source strengths increased to 0.83 μ g/h for BbF, 0.42 μ g/h for BkF, 0.76 μ g/h for BaP, 0.73 μ g/h for BghiP, and 1.2 μ g/h for IcdP. For the non-air-tight stove tests, excluding test D-2, the pollutant source strengths ranged from 1.8 to 49 μ g/h for BbF, 0.88 to 23 μ g/h for BkF, 2.2 to 57 μ g/h for BaP, 1.9 to 53 μ g/h for BghiP, and 2.9 to 86 μ g/h for IcdP.

As previously noted, the penetration factors for individual PAHs fell into two groups when calculated with Eq. 2. The smaller 5-ring compounds (BbF, BkF, and BaP) had lower penetration factors than the 6-ring compounds (BghiP and IcdP), 0.28 + 0.03 compared to 0.78 + 0.11. There are several possible explanations for the existence of two groups of apparent penetration factors for PAHs. First, the smaller compounds could be removed more efficiently by the building shell than the larger compounds. This could be due to differences in relative PAH compostion as a function of particle size. However, other work has shown that relative PAH composition of outdoor particles does not depend on particle size (16-18). difference in indoor reactivity between the two groups of PAH compounds might explain the widely different indoor/outdoor ratios and different apparent penetration factors as calculated by Eq. 2. Published research does not support this arguement either. Two studies indicate that, for outdoor particles, BaP is the most reactive and BkF is the least reactive of the five compounds; although, reactivity with indoor surfaces was not investigated (19,20). A third possible explanation is a difference in volatility that could result in selective "blow off" of 5-ring compounds at the indoor sampling location because of the 20°C temperature difference. Again, research does not support this position. During 18 hours of sampling, we would expect losses of the 5-ring compounds to be less that 10%, based on the work of others (16,21,22). No significant losses are expected for the 6-ring compounds.

Another possible explanation of our observations is entrainment of reentrainment of background dust particles whose origin is not fresh wood smoke. Such particles would not be expected to have the same relative composition of PAHs as wood smoke. As an example, urban aerosols are relatively enriched in 6-ring compounds (23). As discussed earlier, we did observe background particles whose origin was not fresh wood smoke. Arguing against this explanation is the observation that, for the two background tests, the indoor PAH concentrations tracked the outdoor levels. The lack of a satisfactory explanation to account for differences in apparent penetration factors for different PAH compounds does not significantly alter the thrust of this paper. However, we urge caution in the interpretation of results that depend on accurate penetration factors.

Conclusions

All of the wood-burning stoves tested emitted at least trace amounts of CO and suspended particles into the indoor environment based on real-time peaks that corresponded to starting, stoking and/or reloading times. In general, the air-tight stoves emitted only minor amounts of pollution indoors. The one non-air-tight stove was a major indoor pollution source under certain operating conditions. When operated with a large fire, the non-air-tight stove caused levels of TSP to exceed the EPA's 24-hour outdoor air quality standard and caused levels of CO to reach the EPA's short-term standards. Indoor

concentrations of five PAHs greatly exceeded outdoor levels when the non-air-tight stove was used. NO_2 and NO levels were extremely low for all tests while HCHO increased only slightly when the non-air-tight stove was operated. The PAH/TSP ratio was found to be a qualitative indicator of the amount of stove-generated particles contained in the TSP samples. Finally, the stove operating style was found to be an important factor in the indoor emissions from wood-burning stoves.

Acknowledgments

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Table I. Description of Wood-burning Stoves

Stove	Approximate effective fire-box dimensions (cm) ^a (height x width x depth)	Type/Comments
Α	48 x 100 x 46	Air-tight, steel plate construction; can be used as a fireplace
В	32 x 33 x 56	Air-tight, cast iron construction
С	40 x 33 x 65	Air-tight, steel plate construction; brick lined
D ^b	54 x 57 x 36	Non-air-tight, cast iron construction with downstream and upstream dampers; 30-inch Franklin model; can be used as a fire-place

 $^{^{\}rm a}$ Effective dimensions are defined to describe the usable volume for loading firewood if the stove was fully loaded.

 $^{^{\}mbox{\scriptsize b}}$ Stove D is not designed to be fully loaded with wood, but rather to be used with a few logs at a time.

Table II. Summary of CO, submicron particulate, and miscellaneous data from operating a wood-burning stove.

Stove- Test number	Burna time(h)	Fuel consumed (kg)	House air exchange rate (h-1)	(bbm) COp			Ţ	Submicron ^c particles (µg/m ³)				
				Avg.	Peak	Out.	Avg.	Peak	Bkgd.			
"Air-tig	ght"				·							
A-1	14.8	37.3	0.48 ± 0.03	0.7	2.8	0.2	36	140	4.6			
A-2 ^d	8.8	38.4	0.88 <u>+</u> 0.03	0.4	1.2	0.0	25	45	18			
B-1	13.0	21.2	0.42 ± 0.02	1.3	1.6	0.7	15	24	14			
B-2	13.7	27.2	0.44 <u>+</u> 0.02	1.7	2.0	0.7	24	150	15			
C-1	18.5	40.9	0.46 ± 0.03	0.8	1.2	0.7	11	28	11			
C-2	18.0	43.6	0.40 ± 0.02	1.0	1.4	0.6	26	290	17			
C-3	16.1	29.2	0.36 ± 0.01	2.8	3.8	1.1	28	120	6.4			
"Non Air D-1	r-tight" 8.7	24.4	0.67 <u>+</u> 0.03	3.4	7.7	0.0	970	2100	6.3			
D-2	10.4	32.4	0.56 ± 0.07	11	35	0.2	-	-	-			
D-3	5.4	23.2	0.58 ± 0.06	14	43	1.0	1900	10,000	24			
D-4	9.5	38.6	0.66 ± 0.06	1.8	3.5	0.4	210	1900	21			
"No Stor	/e" 17.6		0.30 <u>+</u> .04 ^f		-	- -	3.7	7.	6 -			
Bkgd-2	22.9 ^e		$0.30 \pm .04^{f}$	-	-	-	7.6	19	-			

Burn time is the duration between initial ignition and the last time the fire box temperature dropped below 80°C.

Average concentrations are averaged over rooms and time, peak concentrations are averaged over rooms, and outdoor concentrations are averaged over time.

Livingroom monitored only; particle < 0.6 µm, assuming spherical shape and 2.0 g/cm³ density. Values should be used for relative comparisons and not for their absolute values. "Bkgd." is background indoor concentration before stove start.

d Used as a fireplace.

e Sampling time.

f Based on data during other days under similar wind and temperature conditions.

Table III. Summary of indoor and outdoor TSP and PAH data while operating a wood-burning stove indoors.

•

Stove/ Test	TSP (μg/m ³)	BbF (ng/m ³)		BkF (ng/m ³)		BaP (ng/m³)		BghiP (ng/m ³)		IcdP (ng/m ³)		(Sum PAH)/TSP ^b (ng/µg)	
	In(Δt) ^a Out(Δt) ^a	In	Out	In	Out	In	Out	I n	Out	In	Out	In	Out
Air-tight"													
A-1	71(11.2) 12(11.2)	0.56	0.56	0.31	0.21	0.88	0.47	1.1	0.96	1.7	1.3	0.065	0.30
A-2 ^C	28(19.6) 12(19.6)	3.8	0.52	1.9	0.23	3.5	0.51	3.7	0.59	6.1	1.3	0.69	0.25
B-1	38(6.6) 19(6.6)	0.71	1.5	0.23	0.76	0.94	1.9	. 1.7	1.9	1.7	3.0	0.14	0.47
B-2	49(14.3) 31(14.3)	0.47	1.0	0.20	0.38	0.48	0.83	1.1	1.1	1.8	1.5	0.084	0.15
C-1	24(18.7) 10(18.7)	0.17	0.8	0.07	0.30	0.34	0.55	0.37	0.92	1.1	1.2	0.085	0.36
C-2	34(11.4) 18(11.4)	0.30	1.0	0.13	0.44	0.42	1.1	1.3	1.3	1.5	2.0	0.11	0.33
C-3	39(16.0) 7.2(16.0)	0.73	1.5	0.28	0.63	0.71	1.2	0.92	1.0	1.8	2.0	0.11	0.86
"Non-air-tí	ight"					ŕ							
D-1 ^d Avg.	650(1.0) 14(18.2) 470 (1.0) 420	90 94 69	0.90	30 30 22	0.37	76 41 44	0.61	96 65 60	0.82	120 130 93	1.5	0.63 0.75 0.69	0.31
D-2 ^d Avg.	320(2.3) 11(20.5) 380(1.6) e	27 53 e	0.71	11 23 e	0.34	29 66 e	0.74	23 52 e	0.70	39 84 e	1.2	0.40 0.74 e	0.34
D-3 ^d Avg.	430(2.3) 15(14.2) 820(0.7) 1500	16 420 320	2.0	7.5 200 150	1.2	19 490 370	1.7	18 450 340	1.7	29 740 560	3.9	0.21 2.8 1.2	0.70
D-4 ^d	210(2.0) 27(18.8) 28(12.8)	32 1.4	2.8	16 0.67	1.4	37 2.0	3.1	.35 2.8	3.7	51 3.9	6.0	0.82 0.38	0.62
Avg.	100	11		5.4		13		14		20		0.62	
"No Stove"													
Bk gd-1	24(17.6) 16(17.6)	0.075	0.62	0.059	0.28	0.16	0.68	0.36	0.48	0.45	0.78	0.047	0.18
Bkgd-2	19(22.9) 17(22.9)	0.056	0.20	0.019	0.082	0.044	0.18	0.085	0.14	0.13	0.25	0.018	0.049

At=sampling time in hours.

Sum PAH = BbF + BkF + BaP + BghiP + IcdP.

Used as a fireplace.

The first two indoor TSP and PAH values reported are from two separate samples taken at two different times during the test, the third number is an estimate of the average concentration over the entire burn period using real-time relative particulate concentration data.
Insufficient data to calculate an average value over the entire burn period.

Table IV. Average CO, TSP, and BaP source strengths while stove was operating.

		5		
Test Number	CO (cm ³ /h)	TSPa (mg/h)	BaP ^b (μg/h)	
"Air-tight"		·		
A-1 A-2 B-1 B-2 C-1 C-2 C-3	60 80 60 100 10 40 140	8.7 5.2 3.6 4.5 2.5 3.0 3.8	0.10 0.76 0.05 0.03 0.03 0.02	
"Non-air-tight		`		
D-1 D-2 D-3 D-4	540 1400 1800 220	73 230 16	7.7 57 2.2	
"No stove"				
Bkgd-1 Bkgd-2	c c	1.6 1.1	< 0.01 < 0.01	

Using an outdoor penetration factor of 0.48. Using an outdoor penetration factor of 0.30. a b

Not measured. С

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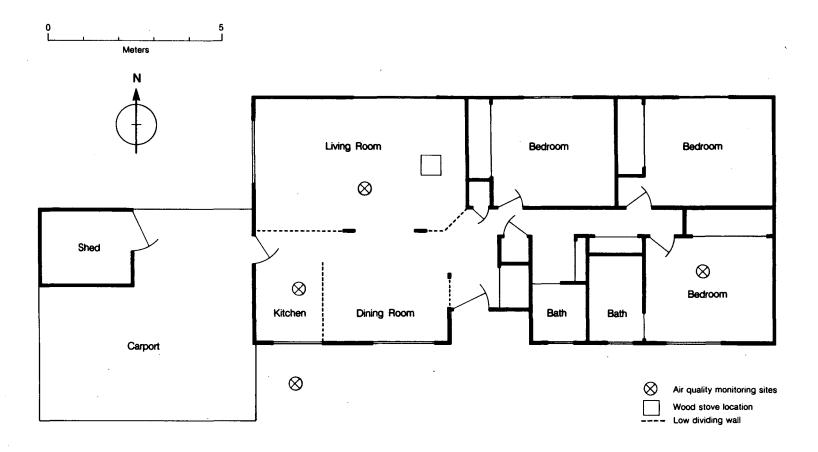


Figure 1. Floor plan of test house.

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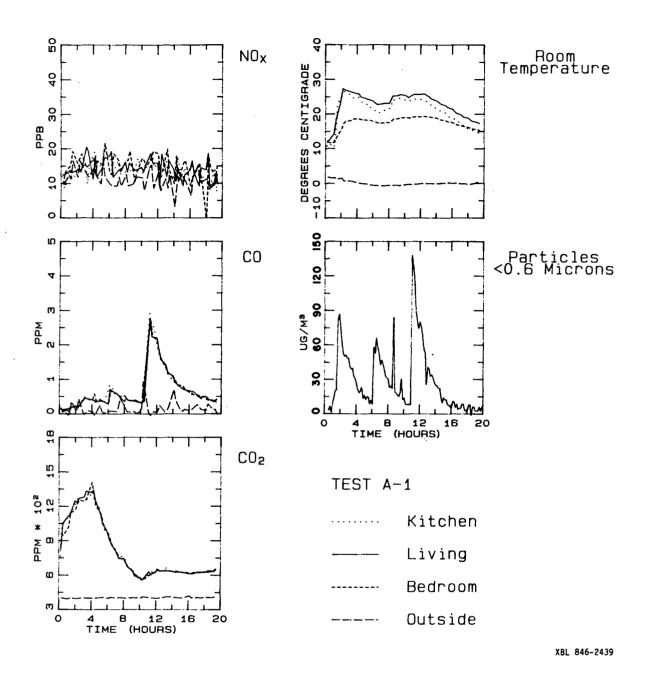
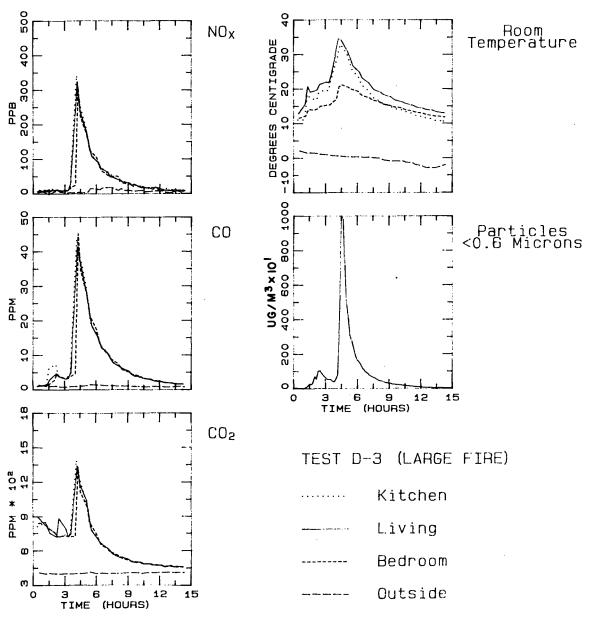


Figure 2. Pollutant concentrations versus time while operating an air-tight stove under normal conditions in a 236 $\rm m^3$ house.



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Figure 3. Indoor pollutant concentrations versus time while operating a non-air-tight stove with a large fire in a 236 m^3 house.

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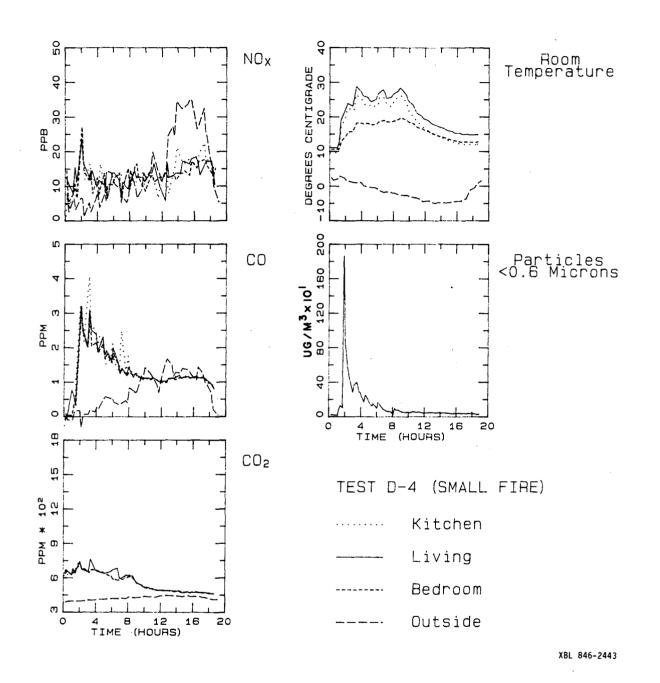


Figure 4. Indoor pollutant concentrations versus time while operating a non-air-tight stove with a small fire in a 236 m^3 house.

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