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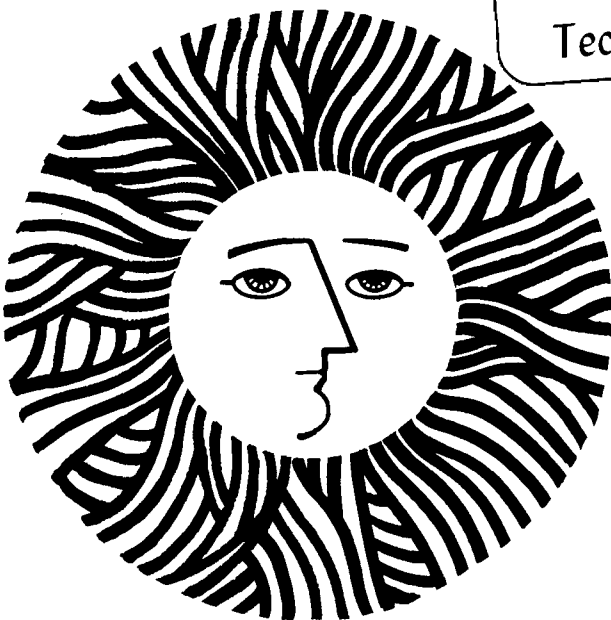
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APPLIANCES AND SIDESTREAM CIGARETTE SMOKE

J.R. Girman, M.G. Apte, G.W. Traynor,
J.R. Allen, and C.D. Hollowell

November 1981

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ABSTRACT

Particulate and gaseous emissions from indoor combustion appliances and smoking can elevate the indoor concentrations of various pollutants. Indoor pollutant concentrations resulting from operating one of several combustion appliances, or from sidestream tobacco smoke, were measured in a 27-m³ environmental chamber under varying ventilation rates. The combustion appliances investigated were gas-fired cooking stoves, unvented kerosene-fired space heaters, and unvented natural gas-fired space heaters. Results showed elevated levels of carbon dioxide, carbon monoxide, nitric oxide, nitrogen dioxide, formaldehyde, and particulates from one or more of the pollutant sources investigated. Our findings suggest that, of the sources examined in this study, nitrogen dioxide and particulates are the most serious contaminants of indoor air, if we use existing standards and guidelines as the criteria. An emission rate model was used to quantify the strengths of the pollutant sources, which are reported in terms of the mass of pollutant emitted per energy unit of fuel consumed (in the case of gas and kerosene appliances) and per mass of tobacco combusted (in the case of smoking).

INTRODUCTION

Indoor combustion appliances and tobacco smoking are primary sources of air pollution in many residences. Gas-fired stoves and unvented space heaters (both kerosene- and natural gas-fired types) emit such potentially harmful pollutants as carbon monoxide (CO), carbon dioxide (CO₂), nitric oxide (NO), nitrogen dioxide (NO₂), sulfur dioxide (SO₂), formaldehyde (HCHO), and respirable particulates; sidestream tobacco smoke contains CO, CO₂, NO, NO₂, respirable particulates and a wide range of organic compounds. The degree of indoor air pollution and, therefore, the degree of health risk to occupants from these sources depends on the type and amount of pollutants entering the occupied space, and the rate of removal processes occurring, e.g., natural infiltration, mechanical ventilation, and chemical reactions.

Calculating emission rates of these combustion-generated pollutants is an essential step in assessing the degree to which these pollutant sources affect indoor air quality. In this paper, we report the pollutant emission rates derived from our studies of a gas-fired stove, gas-fired unvented space heater, kerosene-fired unvented space heater, and sidestream cigarette smoke.

EXPERIMENTAL

Experiments were carried out in an environmental chamber under controlled ventilation conditions; our Mobile Atmospheric Research Laboratory (MARL) was used to measure gas-phase pollutant concentrations.

Both the 27-m³ environmental chamber and the MARL are depicted schematically in Fig. 1. The chamber can be operated under conditions of natural infiltration, typically providing less than half an air change per hour (ach), or under mechanical ventilation, providing higher air exchange rates. Small, variable-speed fans centered on each chamber wall provide mixing of the air to insure uniform pollutant concentrations. When measuring pollutant emission rates from unvented natural gas-fired space heaters a "cold wall" composed of two solar panels helped to remove radiant heat.

The MARL is capable of measuring concentrations of oxygen (O₂), CO, CO₂, NO, and NO₂ from any of three locations within the chamber as well as from the background air surrounding the chamber. Particulate samplers were located inside the chamber. Dew points and temperatures, both inside and outside the chamber, were also recorded. Gas-phase pollutant concentrations were measured at one-minute interval and recorded on magnetic tape; the results were analyzed with a model (Traynor et al., 1981a) to determine emission rates for the various pollutants.

RESULTS

Gas stoves are one of the most studied sources of indoor pollutants (Cote et al., 1974; Himmel and Dewerth, 1974; Traynor et al., 1981a); therefore, sufficient data are available for comparison of their pollutant emissions with those of sidestream cigarette smoke and other combustion appliances. Table 1 presents the pollutants, and their emission rates, from the operation of a gas-fired stove (Traynor et al.,

1981a).

Emission rates of pollutants from an unvented kerosene space heater and from sidestream cigarette smoke are presented in Table 2. The kerosene space heater used in this experiment was a portable convective type that has become very popular recently. The manufacturer's rated output for this heater was 9700 kJ/hr but, based on its rate of kerosene consumption and a heat production value of 36,800 kJ/l, for kerosene, it consumed only 3490 kJ/hr. The heater was operated in the chamber at various ventilation rates. Pollutant concentrations vs time for CO, CO₂, NO, and NO₂ from this heater are shown in Fig. 2 for the ventilation rate of 1.9 ach. The chamber size and the ventilation rate of 1.9 ach may be fairly representative of actual usage conditions; i.e., the manufacturer's literature indicates its ability to heat a small room, assuming some natural ventilation (i.e., from opening a door to adjoining rooms or opening a window). Significant observations from this chamber experiment are that, under these conditions CO and particulate emissions are low, apparently forming the basis for claims that the new-generation kerosene heaters have no harmful emissions (Tutak, 1981). On the other hand, CO₂ emissions are high, concentrations reaching 5000 ppm, the occupational standard established by the U.S. Occupational Safety and Health Administration (U.S. OSHA, 1979), after 45 minutes of operation, and well above the standard of 2500 ppm established by the American Society of Heating, Refrigeration and Air-Conditioning Engineers (ASHRAE, 1981). NO_x emissions are also high, most notably the NO₂ concentration which was greater than 1 ppm over background after 45

minutes of operation. This concentration can be compared to the California short-term (peak one-hour average) outdoor standard of 0.25 ppm (California Administrative Code, Title 17) and the U.S. Environmental Protection Agency's long-term (annual average) outdoor standard of 0.05 ppm (U.S. EPA, 1979), although it did not exceed OSHA's workplace standard of 5.0 ppm (U.S. OSHA, 1979).

The particulate concentrations also shown in Table 2 were from the sidestream smoke of a single cigarette smoked by a volunteer situated inside the environmental chamber. The volunteer, a student, studied in the chamber and smoked at his usual rate. The cigarettes were a popular commercial brand. During this experiment, the ventilation of the chamber was maintained at a low rate. Particulates from the sidestream cigarette smoke were size-fractionated and weighed using a ten-stage piezoelectric cascade impactor. These results, shown in Fig. 3, indicate the very high levels ($390 \mu\text{g}/\text{m}^3$ over background) of total particulate concentrations measured. In addition, 96 percent of this increase involved particulates under $0.4 \pm 0.1 \mu\text{m}$ in diameter--a size range having a high probability of penetrating the pulmonary region of the lung (Task Group on Lung Dynamics, 1966).

In a similar experiment using an automatic smoking machine, CO and particulate concentrations from the non-inhaled fraction of tobacco smoke from a single cigarette were measured. Particulate emissions were similar to those measured previously, and the CO concentration in the chamber peaked at 2.7 ppm above the background.

Figure 4 illustrates typical pollutant concentration profiles for unvented gas-fired space heaters and Table 3 lists their emission rates based on a study currently in progress. These appliances are used principally in the southern and southwestern states. As indicated in Table 3, eight heaters have been tested, ranging in size from 12,700 to 42,200 kJ/hr (12,000 to 40,000 BtuH). O₂ consumption rates are included in this table since there is some concern about its depletion in environments where these heaters are used. The NO_x emission rates from these unvented gas-fired space heaters, which average 12 µg/kJ of N, are lower than those of the kerosene space heater or of the gas-fired stove. On the other hand, fuel consumption rates must also be considered when comparing different appliances. The HCHO emission rates from these unvented heaters averaged 0.81 µg/kJ, which is lower than that associated with gas-fired stoves. CO emissions were found to be much more variable than those of other pollutants. Five heaters had low CO emission rates (less than 30 µg/kJ), while the other three heaters had much higher rates. Even in the same heater, CO emission rates were highly variable during repetitive tests. CO₂ emission rates and O₂ consumption rates were relatively constant from heater to heater and test to test.

The variability of CO emissions prompted a series of tests on the sensitivity of emission rates to adjustments of the air shutter. All unvented gas-fired space heaters had previously been tuned with a portable CO analyzer and inspected visually for flame characteristics. With adjustment of the air shutter as the only variable, tests were run on heater 30D by combusting five ft³ of natural gas in 10 minutes under

identical ventilation conditions (0.4 ach) and measuring peak CO, NO₂ and NO concentrations. The results of these tests are shown in Fig. 5. This plot is similar to an emission vs air/fuel ratio plot with the X-axis representing the percent opening of the air shutter. When the air-shutter opening is increased from 21% to 32%, the CO emissions increase by a factor of nine. This increase of 21% to 32% in the air-shutter opening occurs with less than a 10° rotation of the shutter--a small adjustment that underscores the sensitivity of CO emissions to tuning.

Additional tuning tests were conducted on the 30D to investigate the effect of O₂ depletion on CO emissions. By varying the ventilation rate, steady-state O₂ levels of 20% or 18% could be achieved. At 20% O₂, the steady-state concentration of CO in the chamber could be varied from 4 to 90 ppm by changing the air shutter opening from 0 to 47% open. At 18% O₂, the steady-state concentrations of CO could be varied from 7 to 153 ppm by adjusting the air shutter opening over the same range. This test demonstrated that steady-state O₂ concentrations alone are poor predictors of steady-state CO concentrations.

Conversely, NO_x emissions are not as sensitive to tuning. However, in the excess air regime (air shutter open more than 21%), the NO₂ to NO ratio appears to be extremely sensitive to tuning--increasing from 0.3 at a 21% opening to greater than 300 at a 42% opening. Thus, although NO_x emissions are not very sensitive to tuning, the fraction of NO_x in the form of NO₂ is very dependent upon the state of tune.

This heater, 30D, as previously tuned and tested was one of the highest CO-emitting heaters; after applying the results of tuning curve, however, it was one of the four lowest CO-emitting heaters. Since it is unlikely that extensive tuning of this nature would be routinely performed in the field, all other unvented heaters used for this experiment were left as tuned previously (using a portable CO analyzer and observing flame characteristics).

Three of these heaters (20M, 30M, 40M), all from the same manufacturer, were the lowest CO emitters as well as being among the lowest emitters of HCHO and NO_x. Furthermore, emission rate tests showed that these heaters were relatively insensitive to tuning. This insensitivity is illustrated by the results in Table 4 which compares emission rates from the 30D and 40M heaters under two conditions, one with the air shutter fully opened and one with the shutter fully closed. The heaters from this manufacturer incorporate a very different burner design compared to those of the other two manufacturers. Instead of having many small circular ports in a flat, rectangular burner that produce many small "flamelets," this type of burner has relatively few slots cut across a cylindrically-shaped burner which produce a softer "feathered" flame. It is likely that this burner design accounts for both the lower emission rates and their insensitivity to tuning.

DISCUSSION

While it is impossible to describe all conditions of use of unvented gas-fired space heaters and all environments where they are used, a

simple example illustrates the indoor concentrations of pollutants that might result from their use. Let us assume that the heater is used in a 1400 ft² (130 m²) house with well-mixed interior air; all outdoor pollutant concentrations are zero; the air exchange rate for the house is 1 ach; and the unventilated gas-fired space heater used is a well-tuned 20,000 BtuH heater such that one fourth of the NO_x is NO₂. The heater is operated at full input for a fairly long time (in accordance with manufacturer's recommendation that these particular models be operated only at full input and sized according to house volume and climatic zone). In all cases, the emission rate used is the mean of the eight heaters: for NO₂, 10 µg/kJ; for CO₂, 51,200 µg/kJ; for CO, 34 µg/kJ; and for HCHO, 0.81 µg/kJ. (CO₂ and NO₂ emission rates, as given in Table 3, appeared to follow a normal distribution while the CO emission rates are better characterized by a log-normal distribution, as also observed by Himmel and Dewerth, 1974. Because HCHO is also a product of incomplete combustion, its emission rate is assumed to have a log-normal distribution.) Unlike CO and CO₂, both NO₂ and HCHO are reactive gases and this reactivity would reduce the actual concentrations observed. In an extensively tested research house, NO₂ was observed to have a reactive decay constant of 1.3 hr⁻¹ (Traynor et al., 1981b). The reactive decay constant of HCHO--0.4 hr⁻¹--was measured in our environmental chamber. While it is unknown whether either of these values would apply to other environments, for the purpose of our example we will assume that they do.

With these assumptions and conditions we can proceed. After an hour of continuous operation, the HCHO concentration is 24 ppb, the CO concentration rises above 1 ppm, the NO₂ concentration is 0.139 ppm and the CO₂ concentration is 1200 ppm. Even if the heater were operated continuously under these conditions, the steady-state HCHO concentration increases to only 32 ppb, much less than even the most stringent indoor standard for HCHO, 100 ppb (ASHRAE, 1981). The steady-state CO concentration of 2 ppm is also much lower than EPA's outdoor long-term (8 hr) standard of 9 ppm (U.S. EPA, 1979). The steady-state NO₂ concentration from this well-tuned heater (recalling that NO₂ is only one-fourth of NO_x) is 0.154 ppm, much higher than the EPA long-term (annual) outdoor standard for NO₂ of 0.05 ppm, although it does not exceed the California short-term (one hour) outdoor standard of 0.25 ppm. CO₂, at 1890 ppm, approaches but does not exceed the ASHRAE standard of 2500 ppm (ASHRAE, 1981).

In the above example, note that the pollutant concentrations calculated are specific to the size of the heater, its state of tuning, the length of operation, the volume of the heated space, and the ventilation rate. It is apparent that the potential for CO and HCHO concentrations to reach problem levels in an indoor environment depends very much on the volume of the heated space and the ventilation rate, as well as the heater-specific factors listed above. This is not the case with NO₂; based on existing short- and long-term outdoor standards, NO₂ concentrations are likely to be high under most conditions of operation.

In Table 5, we present a list of specific heaters, both well tuned and poorly tuned, for which we calculated steady-state pollutant concentrations from the emission rates shown in Tables 3 and 4. (The assumptions of a 1400 ft³ house at 1 ach with well-mixed air and appropriate decay constants remained.)

For the well-tuned heaters the results are similar to the previous example, i.e., NO₂ concentrations are high and HCHO concentrations are lower than prescribed by existing standards. For two of these well-tuned heaters, CO concentrations approach the EPA 8-hour outdoor standard of 9 ppm; CO₂ concentrations are high relative to the ASHRAE standard and, as expected, scale with heater input. O₂ levels do not fall below 20.2% (assuming an initial and outside O₂ concentration of 20.9%).

In the case of the poorly tuned heaters, NO₂ concentrations remained largely unchanged, that is, still high. The HCHO concentration is variable; it can be quite high as is in the 30D with an open shutter, but need not be, as is shown by the other HCHO concentrations listed for those heaters. CO concentrations in these heaters are also highly variable and, as shown, can be quite high, approaching U.S. OSHA's 8-hour standard of 50 ppm (U.S. OSHA, 1979), and exceeding EPA's outdoor standards. (The emissions from the 40M heater, as stated previously, are remarkably insensitive to tuning.)

CONCLUSIONS

These data on pollutant emission rates obtained from combustion appliances and sidestream cigarette smoke in an environmental chamber, combined with data on source usage, provide important information for determining the impact of combustion-generated pollutants on indoor air quality under a wide variety of environmental conditions. Other necessary parameters for these determinations are the ventilation rate, reactive decay constants, indoor volume and outdoor pollutant concentration.

Our laboratory studies have shown that unvented combustion appliances and tobacco smoking, in particular, produce enough pollutants to be of concern, both in single-room environments and, based on calculations, in residential-sized buildings. While our findings suggest that both unvented heaters and sidestream cigarette smoke can pose a health risk in spaces where ventilation is reduced, in the case of unvented gas and kerosene space heaters, the NO_2 emissions are high enough to warrant concern even under relatively high ventilation conditions.

The burner design of one manufacturer of an unvented gas-fired space heater appears to reduce pollutant emissions significantly and is insensitive to tuning as well, indicating that improvements in burner design should be pursued. Of course, other techniques for reducing or alleviating the effect of pollutant emissions from these sources should also be developed.

Finally, if we are to determine the degree to which occupants are at risk from exposure to combustion-generated pollutants, we need

information on (1) occupant usage patterns and (2) specific characteristics of the environments where these heaters are used. Such data are not presently available. A survey of this type should be supplemented by field studies to measure pollutant concentrations in a variety of indoor environments and over a wide range of usage patterns.

ACKNOWLEDGEMENT

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Table 1. Pollutant emission rates from a gas stove.

Pollutant	Gas Oven Operated at 180 °C (350 °F) for 1 Hour ^a (µg/kJ)			Top Burners on High Setting with Water Filled Pots (µg/kJ)		
Gases						
CO	227	(213-239) ^b	[2]	201	(172-249)	[4]
CO ₂	43,500	(41,600-46,400)	[3]	45,700	(44,000-47,600)	[3]
NO _x (as N)	6.2	(5.0-6.9)	[6]	8.6	(6.9-10.3)	[4]
NO ^d	6.9			9.3		
NO _x (as N)	10.3			14.1		
SO ₂	0.11	(0.08-0.13)	[5]	1.7	(0.86-2.5)	[2]
HCHO	2.7	(2.4-3.4)	[5]	1.7	(0.86-2.5)	[2]
Particles (< 2.5 µm)						
Carbon				0.22	(0.21-0.23)	[3]
Sulfur (as SO ₄ ^m)				0.01	(0.002-0.02)	[4]
Mass ^e				0.41	(0.24-0.62)	[3]
Mass (< 0.5 µm) ^f	0.015	(0.014-0.015)	[2]	0.51	(0.22-1.00)	[4]
Fuel Consumption	8,370 kJ/hr			9,200 kJ/hr/burner		

^aTime-weighted average of initial and steady-state emission rates.

^bFigures in parentheses represent actual range of emission rates.

^cFigures in brackets represent number of experimental runs.

^dAssuming a volumetric NO₂-to-NO ratio of 1.0.

^eBased on gravimetric analysis of filters.

^fBased on electrical mobility analysis and assuming a particle density of 2.0 g/cm³.

Reproduced from Traynor et al., 1981a.

Table 2. Pollutant emission rates from an unvented kerosene space heater and from sidestream cigarette smoke.

Pollutant	Kerosene-Fired Unvented Space Heater ^a (µg/kJ)	Sidestream Cigarette Smoke ^b (µg/mg)
Gases		
CO	c	130 ^d
CO ₂	151,000	c
NO _x (as N)	36	e
NO ^f	38	e
NO ₂ ^f	60	e
Particulates	c	18
Fuel Consumption	3490 kJ/hr	600 mg/cigarette

^aExpressed as micrograms of pollutant emitted per kiloJoule of fuel consumed.

^bExpressed as micrograms of sidestream pollutant emitted per milligram of tobacco burned; 96% of mass $\leq 0.4 \pm 0.1 \mu\text{m}$.

^cObserved concentration was too close to background levels to calculate an emission rate.

^dDoes not include inhaled fraction.

^ePositive interference from other constituents of sidestream cigarette smoke.

^fAssuming a volumetric NO₂-to-NO ratio of 1.0.

Table 3. Pollutant emission rates from well-tuned unvented gas-fired space heaters operated at full input.

Heater ^a	ach	No. of Tests	CO µg/kJ	CO ₂ µg/kJ	O ₂ µg/kJ	HCHO µg/kJ	N(NO _x) µg/kJ	NO ₂ /NO Volumetric Ratio	Particulates ^b µg/kJ
12D	0.43	3	114	50,000	-67,400	4.20	10	1.46	0.30
20D	0.71	3	29	50,100	-71,700	0.61	14	0.39	0.039
30D ^c	0.59	1	25	49,900	-72,900	0.59	14	0.37	0.006
16A	0.53	1	165	51,500	-71,900	0.55	12	0.85	0.049
40A	0.39	2	63	51,000	-68,900	0.96	14	0.84	0.009
20M	0.38	2	14	50,100	-73,700	0.91	11	0.53	0.079
30M	0.56	1	11	52,600	-73,700	0.43	12	0.34	0.064
40M	0.74	2	13	54,600	-71,300	0.61	12	0.35	0.024

^aFuel consumption in thousands of BtuH, e.g.,
40=40,000 BtuH=42,200 kJ/hr.

^bMass of particulates from 0.0056 to 0.562 µm based upon
electric mobility analysis and assuming a particle density of
2.0 g/cm³.

^cThis heater was subjected to extensive tuning and can be
considered to be optimally tuned.

Table 4. Comparison of pollutant emission rates of two poorly-tuned unvented gas-fired space heaters operated at full input.

Heater ^a	ach	CO µg/kJ	CO ₂ µg/kJ	O ₂ µg/kJ	HCHO µg/kJ	N(NO _x) µg/kJ	NO ₂ /NO Volumetric Ratio
<u>Fully open</u> <u>air shutter</u>							
30D	0.44	517	43,500	-57,900	20.2	3	289
40M	0.37	8	59,100	-59,100	0.49	12	0.31
<u>Fully closed</u> <u>air shutter</u>							
30D	0.41	159	52,500	-70,800	1.35	11	0.65
40M	0.40	35	49,000	-67,200	0.29	10	0.56

^aFuel consumption in thousands of BtuH, e.g.,
40=40,000 BtuH=42,200 kJ/hr.

Table 5. Calculated steady-state pollutant concentrations from specific unvented gas-fired space heaters operating continuously in a 1400 ft² (130 m²) house (1.0 ach) with well mixed air.

Heater	CO ^a mg/m ³ (ppm)	CO ₂ ^a g/m ³ (ppm)	O ₂ %	HCHO ^{a, b} μg/m ³ (ppb)	NO ₂ ^{a, c} μg/m ³ (ppm)
well tuned					
12D	4.6 (4.0)	2.0 (1,100)	20.7	118 (98)	340 (0.18)
16A	8.8 (7.7)	2.7 (1,500)	20.6	21 (17)	420 (0.22)
20M	0.9 (0.8)	3.3 (1,900)	20.5	42 (35)	360 (0.19)
40A	8.4 (7.3)	6.8 (3,800)	20.2	90 (75)	1,200 (0.65)
40M	1.7 (1.5)	7.3 (4,000)	20.2	57 (47)	610 (0.32)
poorly tuned					
30D ^d	52 (45)	4.4 (2,400)	20.5	1,400 (1,200)	430 (0.23)
40M ^d	1.1 (0.9)	7.9 (4,400)	20.5	46 (38)	520 (0.28)
30D ^e	16 (14)	5.3 (2,900)	20.4	78 (65)	610 (0.32)
40M ^e	4.6 (4.1)	6.5 (3,600)	20.2	43 (36)	700 (0.37)

^aBackground concentrations assumed to be zero.

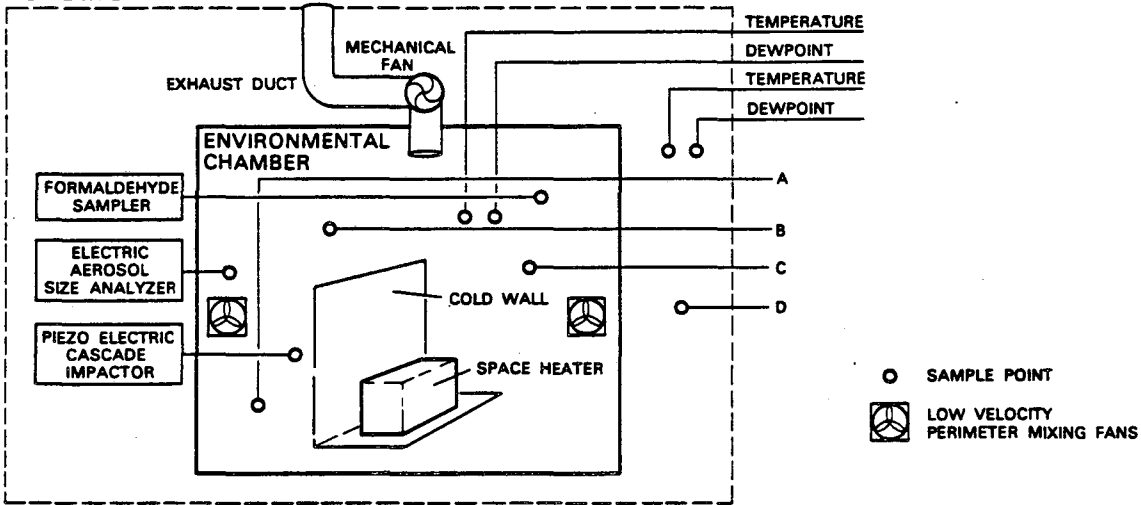
^bA reactive decay constant of 0.4 hr⁻¹ was assumed.

^cA reactive decay constant of 1.3 hr⁻¹ was assumed.

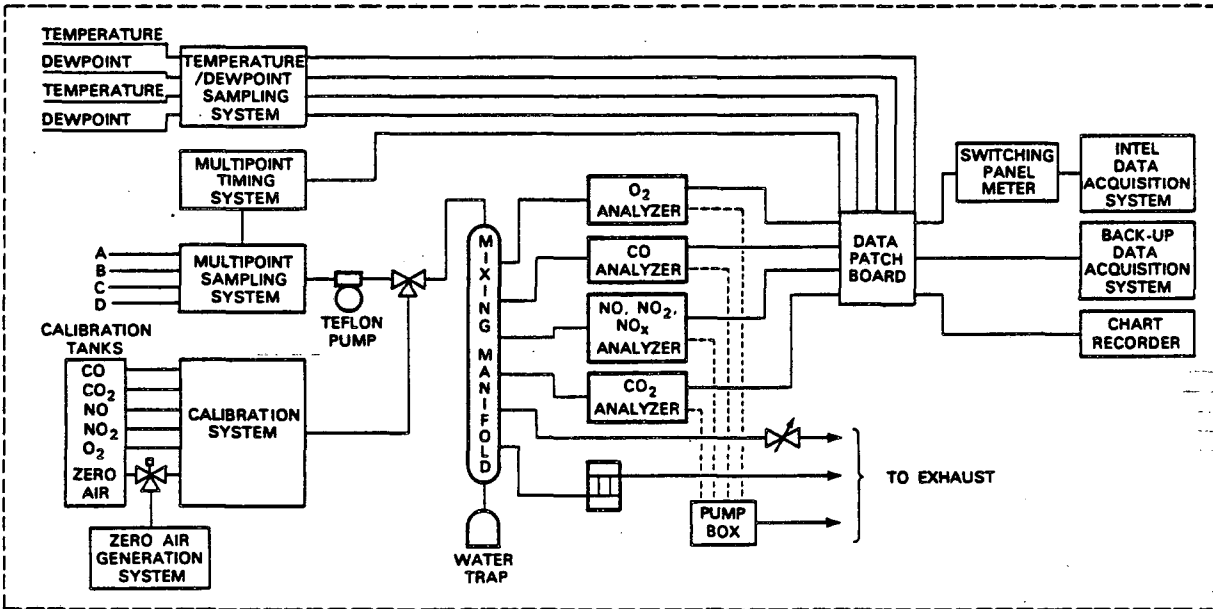
^dFully open shutter.

^eFully closed shutter.

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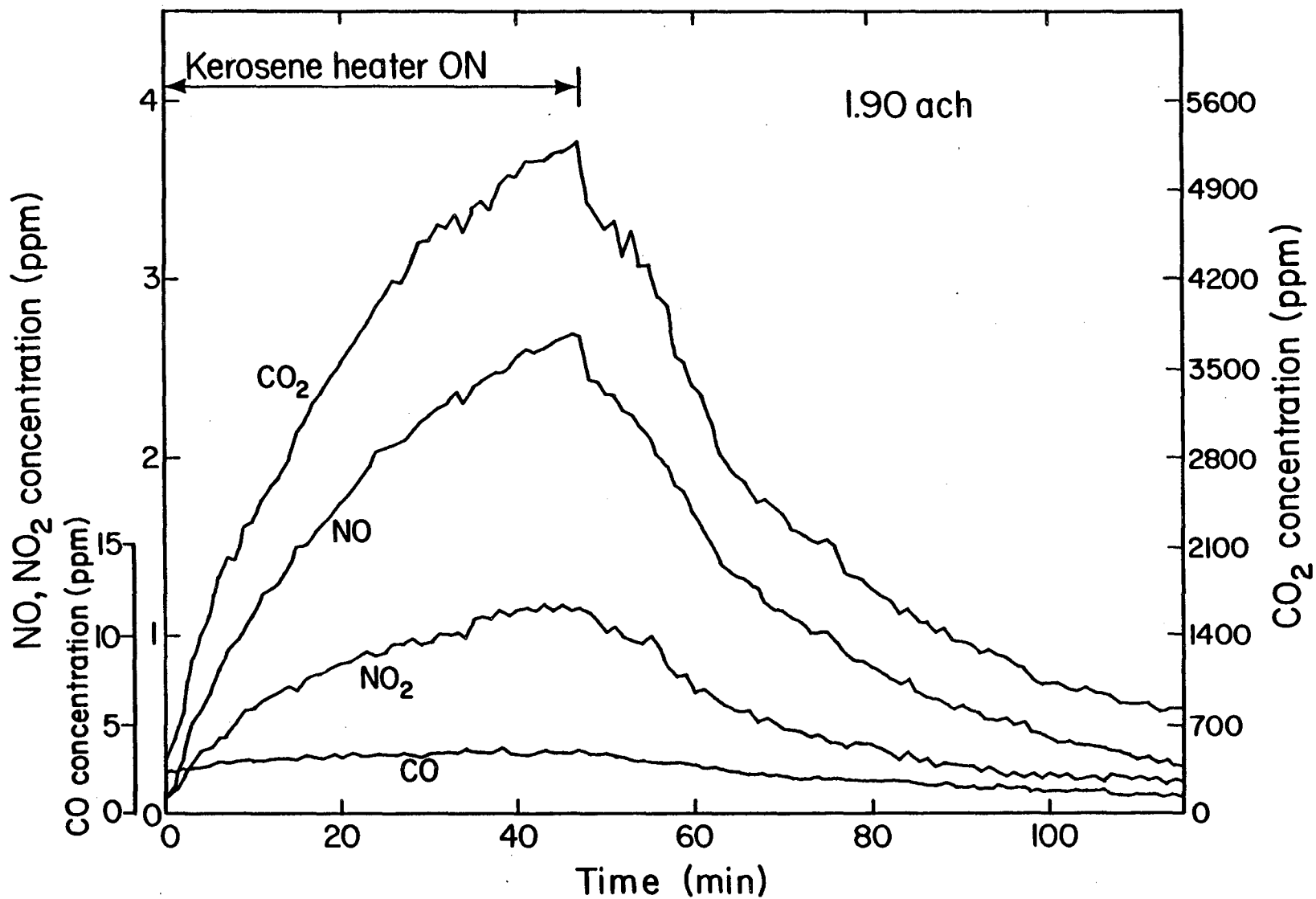


MARL



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Figure 1. Schematic diagram of Lawrence Berkeley Laboratory's Environmental Chamber and Mobile Atmospheric Research Laboratory (MARL).



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Figure 2. CO₂, NO, NO₂ and CO emissions from a portable, convective, unvented kerosene-fired space heater operated for 46 minutes in a 27-m³ environmental chamber with mechanical ventilation (1.90 ach). Particulate emissions were negligible and are not shown.

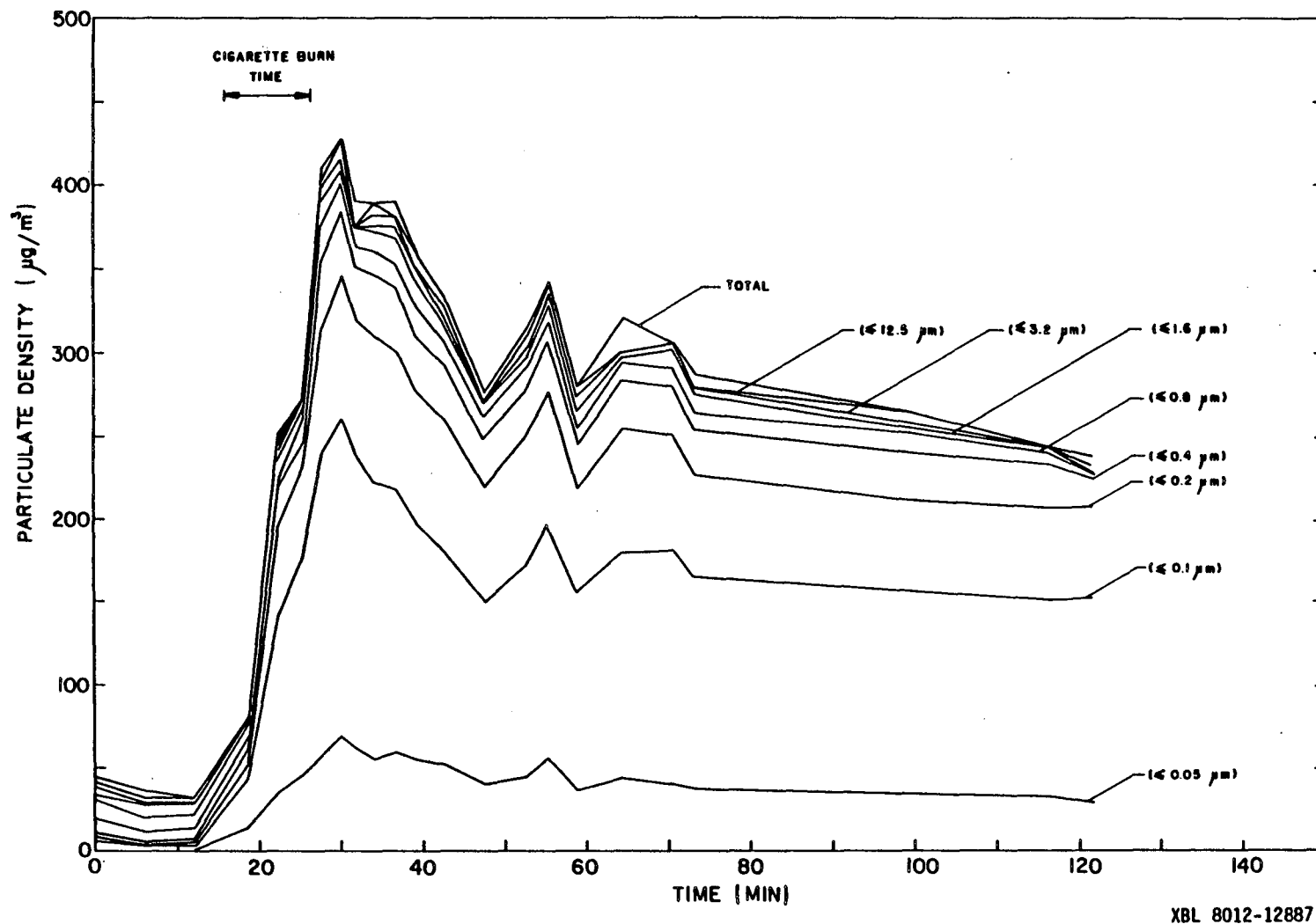
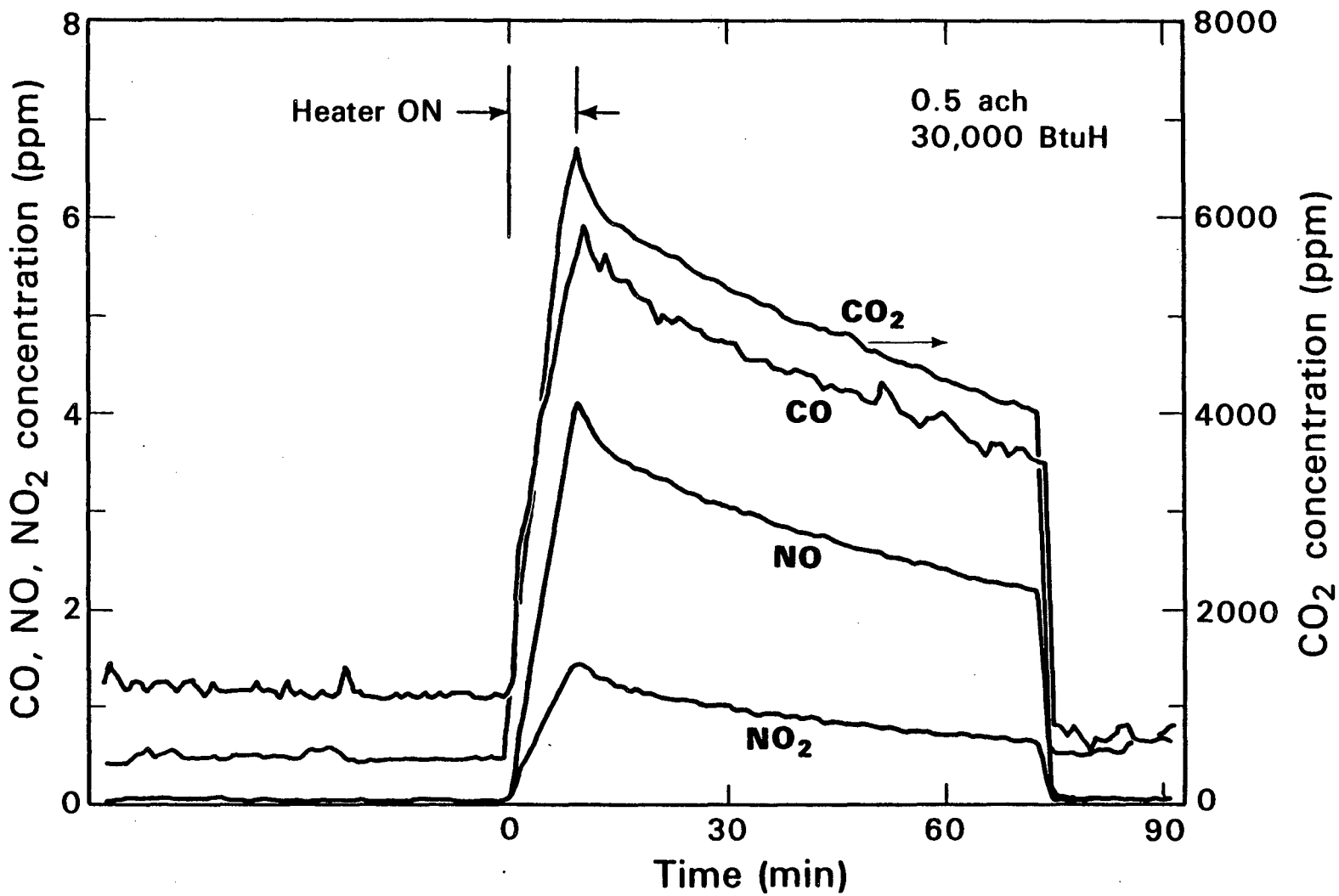
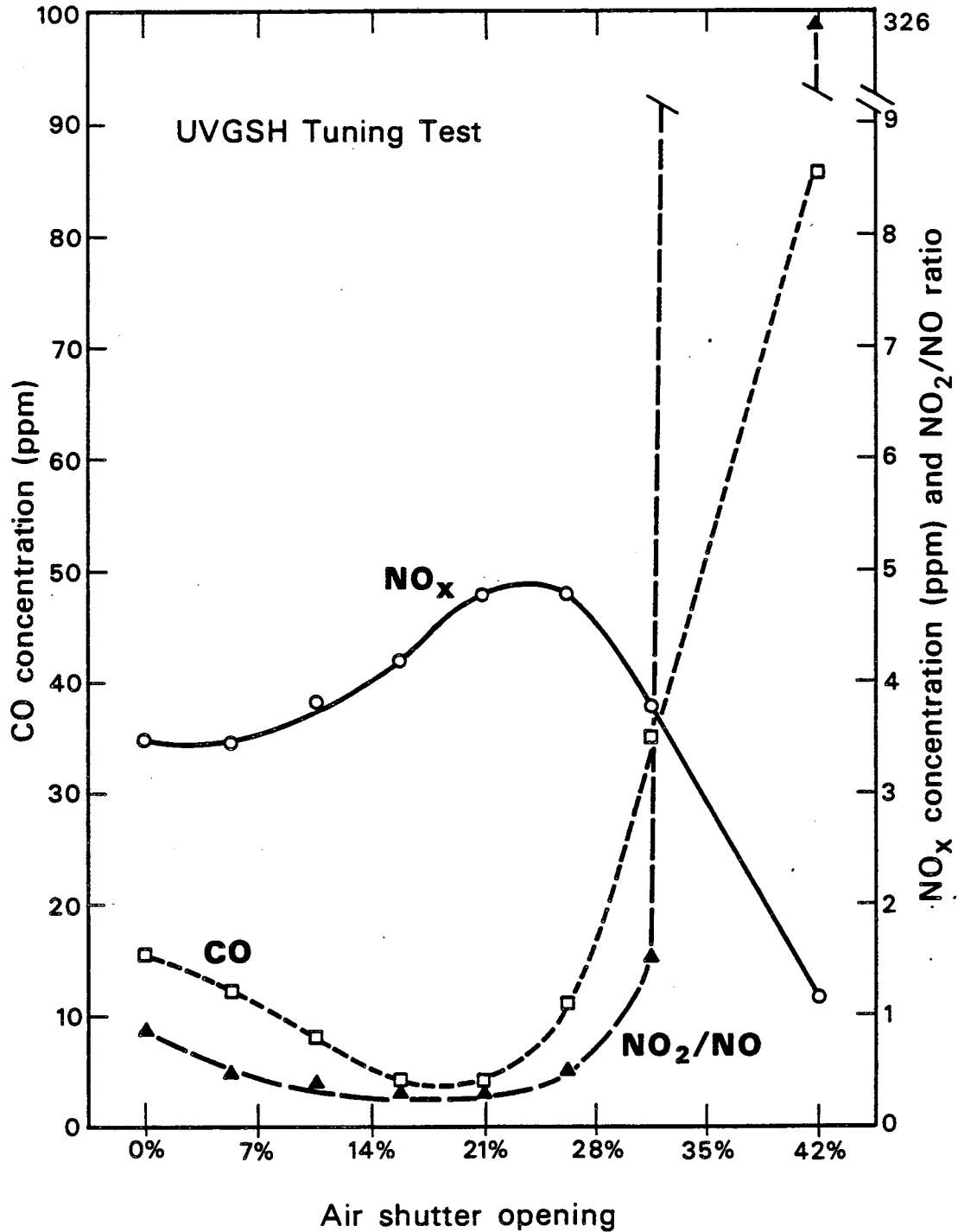


Figure 3. Size-segregated particulate emissions contained in sidestream cigarette smoke from a single cigarette smoked by a volunteer in a 27-m³ environmental chamber without mechanical ventilation (0.3 ach).



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Figure 4. CO₂, CO, NO and NO₂ emissions from a 31,700 kJ/hr (30,000 BtuH) unvented gas-fired space heater operated in a 27-m³ environmental chamber without mechanical ventilation (0.5 ach).



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Figure 5. Peak NO_x and CO concentrations and the NO₂-to-NO volumetric ratios vs. air shutter opening for a 31,700 kJ/hr (30,000 BtuH) unvented gas-fired space heater after combusting 5 ft³ of natural gas in a 27-m³ environmental chamber ventilated at less than 0.5 ach.

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