

Submitted for presentation at the ASHRAE annual meeting,
Los Angeles, California
February 1980

LBL 8394
EEB Env 78-6

An Intercomparison of Tracer Gases
Used for Air Infiltration Measurements*

D. T. Grimsrud and M. H. Sherman
Lawrence Berkeley Laboratory
Berkeley, Ca 94720

J. E. Janssen and A. N. Pearman
Honeywell, Inc.
Minneapolis, Mn 55413

D. T. Harrje
Princeton University
Princeton, NJ 08540

19 April 1979

* The work described in this report was funded by the Office of Buildings and Community Systems, Assistant Secretary for Conservation and Solar Applications of the U.S. Department of Energy under contract No. W-7405-ENG-48.

ABSTRACT

A direct intercomparison has been made between common tracer gases used to measure air infiltration rates in buildings. The results indicate that air exchange rates measured using sulfur hexafluoride, SF_6 , are slightly larger than those measured using methane, CH_4 , or nitrous oxide, N_2O . The ratio of air change rates measured using SF_6 to air change rates measured concurrently using a lighter tracer gas was found to be 1.10 ± 0.10 .

Introduction

The energy cost of excessive air infiltration in buildings has spurred a sharp increase in the number of research projects investigating the magnitude of natural ventilation. The most common technique used to monitor air infiltration rates requires the measurements of the concentration of a tracer gas. The tracer gas, a material easily monitored which is normally not present in the atmosphere, is injected into the space to be tested. When the injection ends the concentration of tracer is measured as a function of time. Outside air, leaking into the test space, replaces the tracer - indoor air mixture which leaks out at the same rate. The rate of change of the concentration of tracer in the indoor air is therefore proportional to the concentration of tracer in the test space, i.e. the concentration decreases exponentially.

This verbal description of the measurement process contains several assumptions about the nature of air infiltration. For example, it assumes that the rate of air infiltration remains constant during the measurement period. In addition, it assumes that the outside infiltrating air mixes uniformly with the indoor air during measurement. In this investigation we examine the assumption that measured air change rates are independent of the type of tracer gas used if adequate mixing of the tracer in the test space occurs.

An ideal tracer gas should meet the following criteria [1]:

It should (1) be inexpensive,

(2) be easily measureable at low concentrations,

(3) be non-toxic and non-allergenic,

(4) be non-flammable,

(5) have approximately the same molecular weight as air,

- (6) not be absorbed on any surfaces within the space under test,
- (7) not be a normal constituent of the air in the test space.

No tracer gas meets all these requirements. Within recent years improvements in instrumentation have permitted measurements of concentration of sulfur hexafluoride, SF_6 , at levels of parts per billion. This is three orders of magnitude smaller than previous techniques which yield measurements in the range of parts per million. The ability to observe such low concentrations makes SF_6 an attractive possibility for use as a tracer gas. However several researchers have expressed the concern that the large molecular weight of SF_6 (146) will cause stratification of the tracer gas after injection. Hunt [1] has discussed the errors which will result if a tracer gas is mixed poorly within the test space. Stratification, which will yield poor mixing, will lead to erroneous infiltration results unless multiple sampling and properly weighted averaging of the tracer from several points occurs.

The experiment described in this paper was designed to make concurrent direct comparisons of air exchange rates measured with different tracer gases in the same test space.

LITERATURE REVIEW

The paper of Hitchin and Wilson [2] is an excellent review of the experimental techniques used in measuring air infiltration. This work has been extended to include work through 1978 by Hunt's careful review of current techniques [1]. Both authors cite previous direct intercomparison results. These are shown in Table 1 below.

Table 1

Published Comparisons between Tracer Gases

<u>Reference</u>	<u>Tracer Gases</u>	<u>Number of Tests</u>	<u>Results*</u>
Warner [3]	Coal Gas, CO ₂	3	1.05±0.18
Collins and Smith [4]	H ₂ , ⁴¹ A	2	0.93±0.01
Howland, et al. [5]	CO ₂ , ⁸⁵ Kr	3	1.00±0.09
Lidwell [6]	N ₂ O, C ₃ H ₆ O	1	0.97
Howard [7]	H ₂ , N ₂ O	many	agreement
" "	O ₂ , N ₂ O	"	"
Hunt and Burch [8]	He, SF ₆	6	1.17±0.14

*The results quoted are the mean values of the ratios of the measured air change rates. The ratio is formed by dividing the air change rate of the heavier gas by the air change rate of the lighter gas.

Warner [3] reported comparisons between coal gas (a mixture of H₂ and CO obtained by passing steam over hot carbon) detected using a Katharometer and CO₂ whose concentration was measured by Haldane gas analysis.

Collins and Smith [4] used the radioactive argon isotope ⁴¹A as a tracer; its concentration was measured with a geiger counter and a ratemeter. A direct comparison was made of the infiltration rate detected using H₂ detected with a katharometer and ⁴¹A; agreement within 8% was seen in two trials.

Howland, Kimber and Littlejohn [5] reported comparisons between air changes measured with the radioactive isotope ^{85}Kr using a geiger counter and a ratemeter. The decay rates were compared with measurements which used CO_2 as the tracer. Its concentration was determined by drawing samples of air periodically and using chemical analysis (the Haldane apparatus) to find the amount of tracer remaining in the test space. Results of 3 tests varied by about 9%.

Lidwell [6] compared results obtained when nitrous oxide, N_2O is compared with acetone, $\text{C}_3\text{H}_6\text{O}$ as a tracer gas. Infrared absorption was used to measure the concentration of N_2O ; acetone concentration was determined by measuring the change in pH which occurred when air containing acetone is absorbed into solutions of hydroxylamine hydrochloride. A single measurement (judged to be accurate to within 10%) produced 3% agreement.

Howard [7] compared N_2O with both H_2 and O_2 . N_2O concentrations were determined with an infrared analyzer, H_2 concentrations with a katharometer and O_2 by absorption in aqueous chromous chloride. Specific results are not quoted in the paper. The author states that close agreement between decay rates using N_2O and O_2 were seen over wide ranges of wind speeds. On the other hand, H_2 decay rates were substantially higher than N_2O . The evidence suggested that diffusion of H_2 through the walls of the unpainted gypsum of the test space was the source of the discrepancy. This hypothesis was tested by repeating the tests after the walls were sealed with two coats of latex paint and also repeating the tests in a laboratory with masonry walls. The discrepancy was not present in the latter two sets of tests.

Hunt and Burch [8] compared air change rates using He and SF₆ as tracer gases to examine the influence of molecular diffusion on the infiltration process. Their test space was a four-bedroom townhouse constructed within an environmental test chamber. If molecular diffusion were important in the infiltration process, the air change rate measured with He would be significantly larger than that measured with SF₆. In fact, slightly larger air change rates were seen when SF₆ was used as the tracer rather than He. Six trials were made. The ratio of the air change rate measured with SF₆ to that measured with He was 1.17 with a standard deviation of 0.14.

TEST SPACE

The test reported in this paper were conducted in a one story unoccupied residence in Walnut Creek, California. This house is leased by the Lawrence Berkeley Laboratory as a research facility for the Energy Efficient Buildings program. A plan view of the house and its surroundings is shown in Fig. 1. The floor plan of the house is shown in Fig. 2. The volume of the living space of the house is 230 m³, its floor area is 100 m² and the area of the six surfaces bounding the living space is 300 m².

The windows of the house are single glazed aluminum sliding windows with rubber weatherstripping. The house has two wooden exterior doors and a single sliding glass patio door. Prior to these tests, one of the doors had been removed and had been replaced by a plywood panel upon which a fan was mounted for air leakage tests.

The house is heated with a forced air gas-fired furnace system, A central air conditioning system uses the same duct system. The return duct opening, located in the ceiling of the hallway leading to the bedrooms, and the supply duct diffusers located on the floor throughout the house, are both shown in Fig. 2,

TEST PROCEDURE

All tests in this intercomparison used a tracer gas concentration decay technique to measure the air exchange rate of the test space.

During all tests except test 2 and 9 (cf. Table 3, below) the doors and windows were closed and the furnace was off. During tests 8 and 9 windows were open 1 cm. The furnace blower, however, ran continuously to provide mixing for the injection of the tracer gas and to provide a suitable location for sampling the air-tracer gas mixture. The blower capacity of the furnace fan was measured and was found to be $0.4 \text{ m}^3/\text{s}$; this is equivalent to a volume flow rate of 6 house volumes per hour.

Indoor temperature and relative humidity were measured using a hygro-thermograph located in the living room. Outdoor weather conditions, dry bulb temperature, wind speed and direction, were measured using a weather tower mounted on the roof of the garage (cf. Fig. 1).

The fireplace chimney and the stove vent in the kitchen were both covered with plastic and taped closed.

The groups conducting the tests, and the tracer gases used are shown in Table 2.

Table 2

<u>Research Group</u>	<u>Tracer Gas</u>	<u>Detector</u>
Honeywell	CH ₄	IR Analyzer
Princeton	SF ₆	Electron Capture
LBL	N ₂ O	IR Analyzer
LBL	C ₂ H ₆	IR Analyzer
LBL	SF ₆	Electron Capture

Interference between CH₄ and N₂O prevented simultaneous measurements of tracer gas concentrations using these gases; therefore the tests were organized using the schedule shown in Table 3.

Table 3

<u>Date</u>	<u>Test Number</u>	<u>N₂O</u>	<u>SF₆</u>	<u>CH₄</u>	<u>C₂H₆</u>
5/9/78	1	x			
5/9	2			x	
5/9	3	x	x(P) [*]		
5/9	4		x(P)	x	
5/9	5	x	x(B)		
5/10	6	x	x(B)		
5/10	7		x(B)	x	
5/10	8		x(B)	x	
5/10	9	x	x(B)		
5/10	10		x(B)	x	
5/10	11	x	x(B)		
5/11	12	x	x(B)		x
5/12	13	x	x(B)		
5/12	14	x	x(B)		

* Test 3 and 4 using SF₆ were analyzed at Princeton (indicated by (P)) while the SF₆ tests 5 through 14 were analyzed at Berkeley (indicated by (B)).

The samples analyzed at Princeton were collected at the test site in aluminized mylar sample bags which were then shipped from California to New Jersey.

ANALYSIS PROCEDURE

After injection, the rate of change of tracer gas concentration is the product of the air change rate, A, and the concentration in the test space.

$$\frac{dC}{dt} = -AC \quad (1)$$

If the air change rate is constant the solution of eq 1 is simply

$$C(t) = C_0 e^{-At} \quad (2)$$

where C₀ is the concentration of tracer at time t = 0.

Several procedures can be used to analyze data relating concentration and time. In these investigations, concentration was plotted as a function of time using semilog graph paper.

Three SF₆ decays, trials 6, 11 and 14 were rejected after examining the graphs. Either non-uniform mixing, non constant decay rates or instrumentation malfunctions caused this behavior.

EXPERIMENTAL RESULTS

Table 4 shows the results of the measurements.

Test Number	Air Change Rates			Wind Speed m/s	Δt °C	Ratio*
	N_2O	CH_4	SF_6			
	(hr ⁻¹)	(hr ⁻¹)	(hr ⁻¹)			
1	0.49	-	-	4	1	--
2	-	0.75	-	5	-3	--
3	0.64	-	0.71 (P)	4	-2	1.11
4	-	0.63	0.76 (P)	6	-4	1.21
5	0.69	-	0.76 (B)	7	7	1.10
6	0.91	-	--	8	6	--
7	-	0.89	0.94 (B)	8	5	1.06
8	-	1.27	1.59 (B)	9	4	1.25
9	1.25	-	1.19 (B)	9	2	0.95
10	-	0.72	0.80 (B)	7	3	1.11
11	0.51	-	--	4	6	--
12**	0.61	-	0.66 (B)	6	-1	1.08
13	0.58	-	0.70 (B)	3	-3	1.21
14	0.47	--	--	4	-6	--

*The ratio quoted is the air change rate of SF_6 divided by the air change rate of the lighter tracer gas.

**During test 12 ethane, C_2H_6 , was also used as a tracer gas. The air exchange rate measured was 0.68 hr⁻¹; this yields a ratio of 0.97.

The mean value of the ratio for all the tests was 1.10 ± 0.10 . The mean value for the comparisons of SF_6 with N_2O was 1.09 ± 0.09 while the value for the comparisons with CH_4 was 1.16 ± 0.09 .

The uncertainties listed with each of the ratios is the standard deviation of a single measurement. The t distribution with 9 degrees of freedom was used to calculate the expected range of the ratio. This result predicts that the actual range of the ratio, \bar{r} , lies within the range

$$1.01 \leq \bar{r} \leq 1.20$$

at the 99% level of confidence [9]. The t distribution assumes sampling from an infinite sample in which the scatter of results is only due to random effects. Our results, therefore, suggest that:

(a) Systematic errors exist in the measurement procedure which results in SF_6 concentration decays that are too large or N_2O and CH_4 decays which are too small; or

(b) Sulfur hexafluoride overestimates the "true" air infiltration rate when used as a tracer gas.

We remind the reader that the results above refer to two independent sets of measurements of the concentration decay of SF_6 were made using equipment at Princeton and LBL.

A source of additional information to help resolve this problem is contained in the paper of Hunt and Burch [8]. As discussed above, these authors compared tracer gas measurements of infiltration rates using SF_6 and He. If we include their measurements with the measurements we have described we obtain a ratio $A_{\text{SF}_6} / A_{\text{lighter gas}}$ of 1.13 ± 0.12 . Again if we use the t-distribution with 15 degrees of freedom we predict that the actual ratio, \bar{r} , will lie within the range

$$1.04 \leq \bar{r} \leq 1.22$$

with 99% confidence.

Adding additional independent measurements to the sample of measurements described in this report did not increase the likelihood that the discrepancy seen is the result of systematic errors in the measurements. Rather they support hypothesis (b).

The results show that a difference exists between air exchange rates measured using SF_6 and air exchange rates measured using lighter tracer gases. The difference, however, is small and represents the range of uncertainty which we estimate is present in any tracer gas measurement (5% to 10%). An example of this is test 12 in the measurements reported above. The air exchange rates measured using C_2H_6 , SF_6 and N_2O were 0.68 hr^{-1} , 0.66 hr^{-1} , and 0.61 hr^{-1} . These values represent the range of values seen whenever air exchange rates are measured; consequently the differences seen in this intercomparison are scarcely large enough to be significant.

We have examined the measurement process for physical effects which would bias the data in the direction observed. On this basis two effects, molecular diffusion and absorption of N_2O by water vapor, can be eliminated immediately.

If molecular diffusion were important in air infiltration, air change rates measured with light gases which have larger thermal speeds would be larger than those measured with heavy gases. This is the opposite of what we have observed.

If a significant amount of N_2O were absorbed by water vapor in the test space, air exchange rates measured with N_2O would tend to be larger than those using SF_6 . Again the opposite result was actually seen.

Another possibility to consider is settling of the tracer gas in the test space. Since the tracer is sampled at the return duct of the furnace, which is located on the ceiling in the test space, settling of the heavy gases

during the course of the measurement would appear to increase the air exchange rate measured using a heavy tracer gas such as SF_6 .

However, a simple calculation will show that this is quite unlikely. The tracer gases were injected into the return duct of a forced air heating system and are well mixed after a short time. Other results [10] allow us to quote a mixing time of the order of 5 minutes for this house. Therefore after 5 minutes the tracer gas is well mixed throughout the test space -- and this mixing continues throughout the concentration decay measurement,

Buoyancy effects occur on the macroscopic, not microscopic, level. The fractional difference in density between a macroscopic volume of gas containing air and one containing 1 ppb (part per billion) SF_6 is 4×10^{-9} . Since the acceleration due to buoyant forces is $\frac{\Delta \rho}{\rho} g$, the effective acceleration of our volume element containing SF_6 is $4 \times 10^{-10} g$. It would take about 3 hours for such an element to settle 2 meters in still air under an acceleration of that magnitude. However, since the furnace blower moves 6 volumes of house air through it each hour, forced mixing and convective mixing certainly dominate buoyancy effects. We conclude that stratification due to the heavy SF_6 molecule is unlikely after the gas is initially mixed with room air.

Other possibilities exist. Rather than comparing tracer gases we may in fact be comparing instrumentation systems. The SF_6 is detected with electron capture gas chromatographic techniques while the concentrations of the lighter gases are measured using infrared absorption techniques or the change in thermal conductivity of helium-air mixtures.

Another possibility may be absorption of the SF_6 on some material present in the house. A small absorption rate could have a significant effect on measurements in the parts per billion range while they would not be noticed in measurements of concentrations of parts per million.

We must be careful to emphasize, after this extensive discussion, that we are searching for an explanation to a discrepancy that is typical of the uncertainties seen in infiltration measurements. While the difference may be real, it should not preclude use of one gas in preference for another when choosing a tracer gas.

CONCLUSIONS

A direct comparison of air change rates measured using SF_6 and either N_2O or CH_4 shows that SF_6 gives a slightly larger value than the lighter gases. The difference cannot be explained using ideas of molecular diffusion. Stratification due to the settling of the heavier tracer gas seems to be an unlikely cause of the effect. While the difference appears to be real, it is small enough so that it is likely unnoticed in the uncertainly associated with a single tracer gas measurement.

ACKNOWLEDGEMENTS

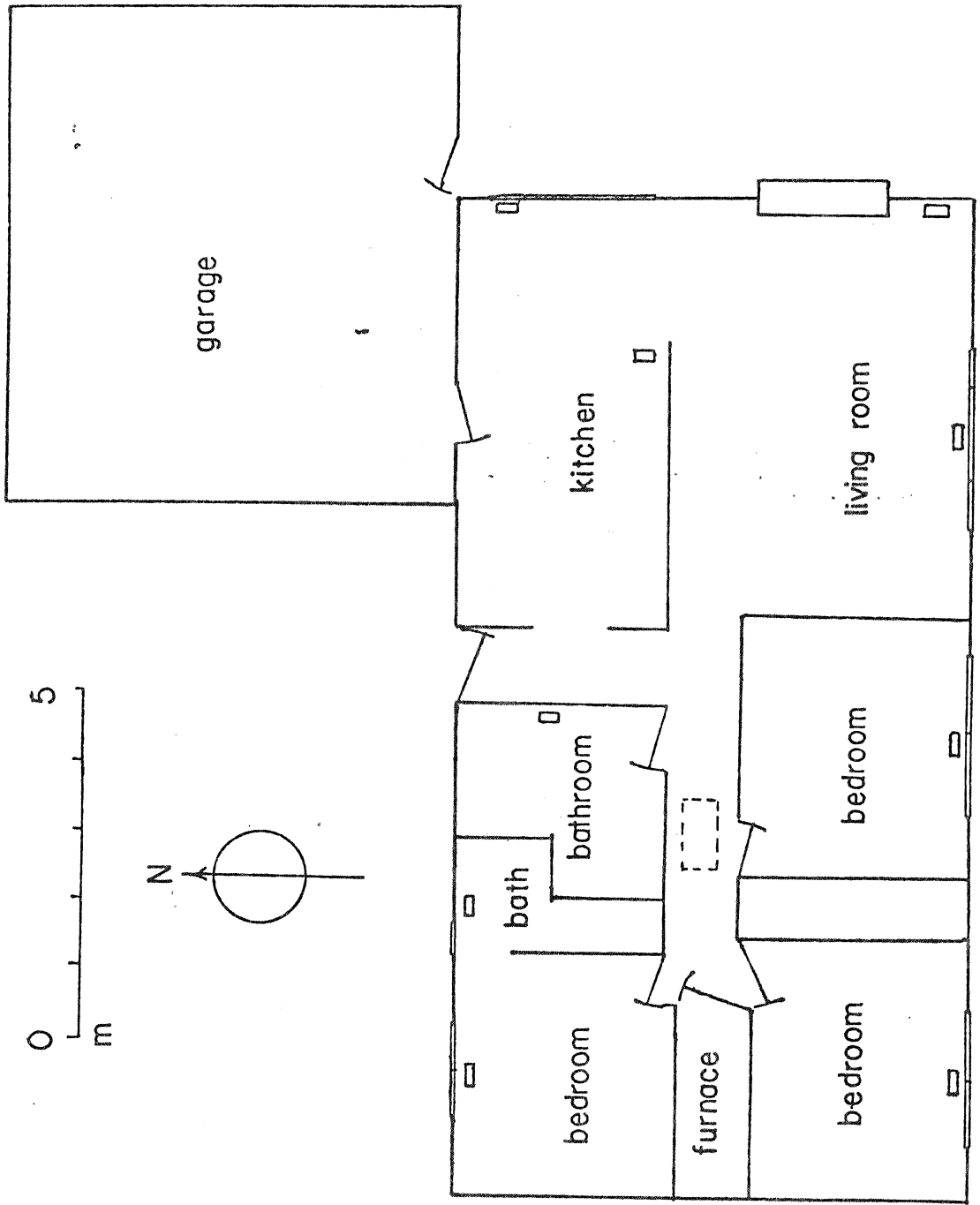
The authors express their appreciation to Rick Diamond and Ron Kammerud of LBL and Howard Ross of the Department of Energy for their support during this project.

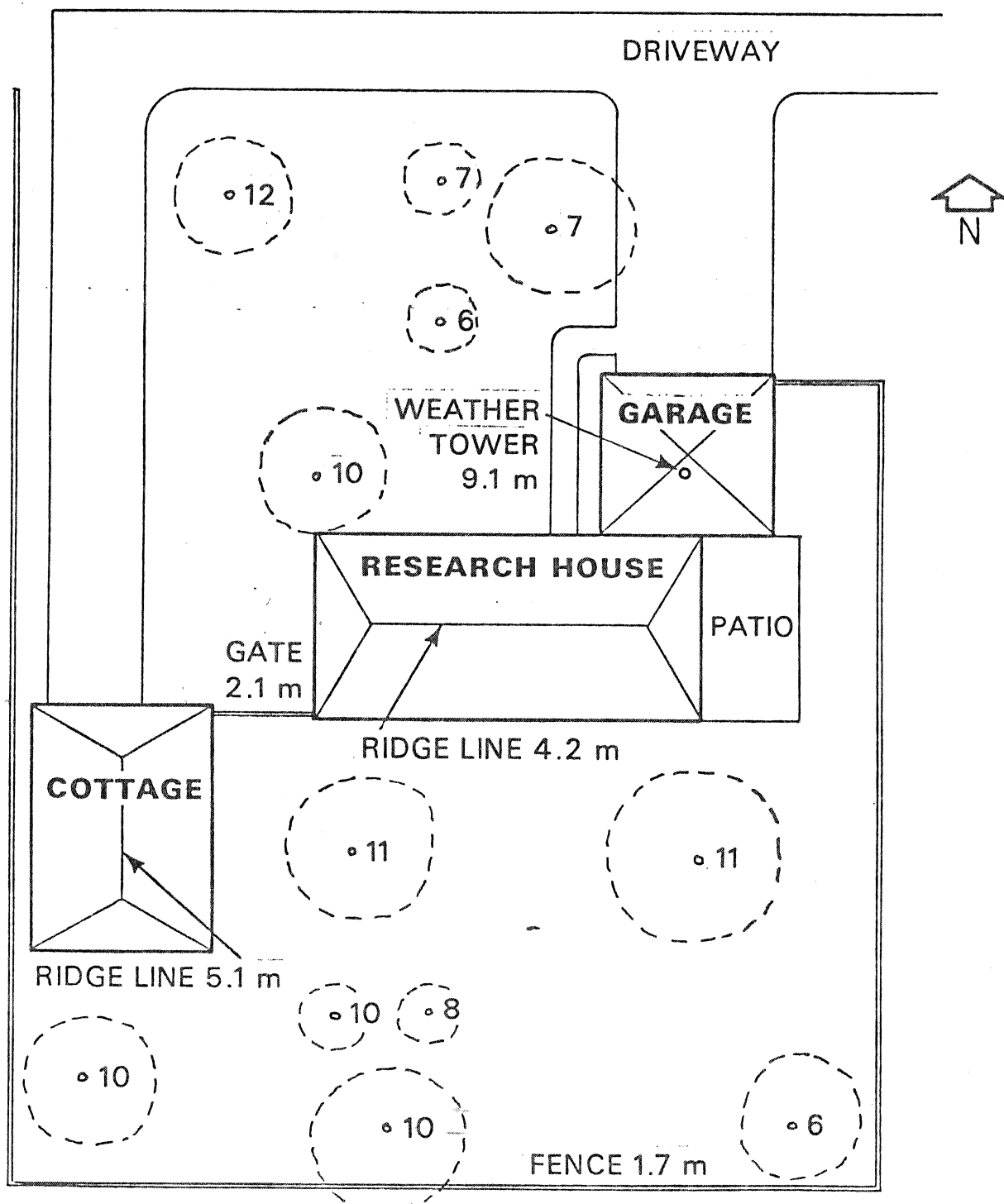
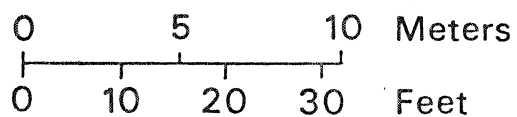
REFERENCES

- [1] Hunt, C. M. (1978), "Air Infiltration: A Review of Some Existing Measurement Techniques and Data", NBS Report, To be published in the Proceedings of the Symposium on Air Infiltration and Air Change Measurements, ASTM, Washington, D.C., 13 March 1978.
- [2] Hitchin, E. R., Wilson, C. B. (1967), "Review of Experimental Techniques for the Investigation of Natural Ventilation in Buildings", Build. Sci. 2: 59-82.
- [3] Warner, C. G. (1940), "Measurements of the Ventilation of Dwellings", J. Hyg. 40: 125-153.
- [4] Collins, B. G., Smith, D. B. ([1955]), "Measurements of Ventilation Rates Using a Radioactive Tracer", J. Inst. Heat Vent. Eng. 23, 270-274.
- [5] Howland, A. H., Kimber, D. E., Littlejohn, R. F. (1960), "Measurements of Air Movements in a House Using a Radioactive Tracer Gas", J. Inst. Heat. Vent. Eng. 28: 57-71.
- [6] Lidwell, C. M. (1960), "The Evaluation of Ventilation", J. Hyg. 58: 297-305.
- [7] Howard, J. S. (1966), "Ventilation Measurements in Houses and the Influence of Wall Ventilators", Build. Sci. 1: 251-257.
- [8] Hunt, C. M., Burch, D. M. (1975) "Air Infiltration Measurements in a Four-Bedroom Townhouse Using Sulfur Hexafluoride as a Tracer Gas", ASHRAE Trans. 81: 186-201.
- [9] Freund, J. E. (1971) Mathematical Statistics, 2nd ed. Englewood Cliffs, N. J.: Prentice-Hall Inc.
- [10] Condon, P. E., Grimsrud, D. T., Sherman, M. H., Kammerud, R. C. (1978), "An Automated Controlled-Flow Air Infiltration Measurement System", LBL Report 6849, to be published in the Proceedings of the Symposium on Air Infiltration and Air Change Rate Measurements, ASTM, Washington, D.C., 13 March 1978.

FIGURE CAPTIONS

- Fig. 1 Plan view of the Walnut Creek Research House and its surroundings.
- Fig. 2 Floor plan of the Walnut Creek House. The return duct is located as shown in the hallway ceiling; the supply duct diffusers are located on the floor.





(Numbers are height of trees in meters)

XBL 788-1537