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Application of Tracer Gas Analysis to Industrial Hygiene Investigations.

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1 Introduction

This article discusses the application of tracer gas methods to industrial hygiene investigations. It introduces the basic concepts necessary to understand the application of tracer gas methods to particular airflow and contaminant movement measurements. It provides an overview of existing methods which can be used to obtain quantitative data on a variety of airflow and contaminant movement related questions which often are of interest to the industrial hygienist. A general description of each method is given, along with the pertinent equations used to extract quantitative information from the test results. Selected results from the use of each method are summarized. It will discuss the use of tracer methods to evaluate building and industrial ventilation systems and methods currently being developed to measure ventilation effectiveness. It will also discuss how to use ventilation measurement along with contaminant concentration measurements to infer contaminant source strengths. It will review the methods used to measure the performance of various systems designed to contain an airborne hazardous substance and assess the potential for spread of this substance into non-containment areas. The topics treated are the evaluation of fume hood re-circulation and re-entrainment, the evaluation of re-entrainment by building air intakes, the simulation of hazardous releases and the performance of secondary containment ventilated enclosures using tracer gases.

Tracer gas methods have been used for

- measuring building ventilation and air infiltration rates
- measuring airflow rates in ducts
- testing fume hoods for re-circulation and re-entrainment
- measuring exhaust re-circulation and re-entrainment
- estimating contaminant source strengths
- evaluating performance of ventilation systems and ventilation effectiveness
- characterizating hazardous substance releases
- evaluating temporary safe havens
- determining flow patterns and flow path tracing
- evaluating airborne hazardous substance containment
- validating models which predict airflow and contaminant levels.

Tracer gas methods have been used in office buildings, hospitals, schools, laboratories, chemical facilities, nuclear power plants, military facilities, semi-conductor fabrication facilities, pulp and paper mills, pharmaceutical facilities and petrochemical plants. In most applications, tracer gas methods are analytical tools which provide accurate means of determining airflow related parameters which are useful to industrial hygienists and in many situations are the only means of obtaining quantitative information. However, these methods are not yet widely known or used outside the research community.

1.1 Characteristics of a Desirable Tracer Gas

Though in principle any gaseous substance not normally found in the area being measured can be used as a tracer gas, the desirable properties of a substance used as a tracer in occupied buildings are that it be

- non-toxic and non-allergenic at the levels used
- chemically inert, odorless and tasteless
- non-flammable and non-explosive
- not naturally present in the building nor as a normal constituent of air
- easily transported and dispersed as an atmospheric gas
- easily and economically measured with high reliability
- measurable by established experimental techniques which preclude interference with air

• commercially available.

Historically, gases such as helium, hydrogen, methane, carbon dioxide, carbon monoxide, nitrous oxide and even radioactive gases have been used as tracers for ventilation and airflow studies. These gases obviously do not have all the properties of an ideal tracer.

1.2 Physical Properties of a Tracer Gas

Currently, the most commonly used tracers are sulfur hexafluoride, halogenated refrigerant compounds and perfluorocarbons. These gases are electronegative tracer gases which can be detected by electron capture gas chromatographs. They have most of the properties of an ideal tracer. They can be detected from the parts per million range to the parts per trillion range. There are several methods which can be used to measure tracer concentrations. These are listed in table 1. In selecting a tracer gas detector and designing one's measurement procedure several characteristics of the detector need to be considered including the range of detectable concentrations, the device's analysis time, the required airflow rate of the sampling device, and the need for expendable supplies such as compressed gas or other chemicals. Table 2 gives the relative costs for several tracer gases. As can be seen by examining the data in table 2, very large volumes can be measured at reasonable cost if gas detection equipment with measurement capabilities in the ppb or ppt range are used. Note that the column labelled "Detectable Concentration" should not be interpreted as "minimum detectable concentrations" as reported by some manufacturers but as the value at which accurate measurements (5% or less) can be made . The authors have successfully performed ventilation tests in buildings which have volumes over 10 million cubic feet. Table 3 lists the currently used tracer gases which can be used to measure airflows in large buildings at modest costs by using small quantities of tracer.

For tracer studies in commercial and industrial buildings, the ability to measure in the parts per billion range (and below) is very convenient: only small quantities of tracer need to be used, these levels are well above typical inside and outside backgrounds (with the exception of those halogenated compounds used as refrigerants), extreme care in handling of the gases is not required, and the levels are at least ten thousand times lower than accepted OSHA standard environmental levels for the respective gases.

2 The Evaluation of Ventilation Systems Using Tracer Gas Methods

This section will discuss the various tracer methods used to evaluate building ventilation systems and methods currently being developed to measure ventilation effectiveness. It will also discuss how to use ventilation measurement along with contaminant concentration measurements to infer contaminant source strengths.

In the evaluation of the performance of ventilation systems, tracer gas methods can be used for

- measuring building ventilation and air infiltration rates
- measuring airflow rates in ducts
- measuring the percentage of outdoor air supplied to the building
- measuring ability of a ventilation system to remove contaminants
- estimating contaminant source strengths
- evaluating ventilation effectiveness

The evaluation of ventilation systems in large buildings will be treated. A brief description on how to use tracer gases to measure airflows in ventilation systems, the assessment of ventilation effectiveness and the use of tracer gases in the estimation of contaminant source

strengths will be given. The inaccuracy of using CO_2 measurements for measuring ventilation rates will be discussed. Finally some examples of using multiple tracers gases for the assessment of complex airflows will be presented.

2.1 Measurement of Ventilation and Air Infiltration

Determination of the ventilation characteristics of residential, industrial and commercial structures has developed into a relatively well-known technology over the last fifteen years. Concerns of health, safety and energy conservation have largely been responsible for driving this development. Tracer gas characterization of ventilation systems has become widely accepted within the building engineering community. In fact ASTM Standard E-741 was promulgated to provide a standard method for measuring air-leakage (ventilation) rates within structures. Over ten years ago the International Energy Agency established a center for air infiltration and ventilation in the United Kingdom to which the United States is a member. This organization publishes a quarterly review of air infiltration and ventilation research and maintains a data base on air infiltration and ventilation research, publications and measurement techniques.

There are three principal techniques for quantifying the air leakage/ ventilation rates within a structure: the tracer decay method, the constant injection method and the constant concentration method. The fundamental principle underlying each of these techniques is the mass balance equation describing the release of a tracer gas in a building (see Appendix I). The critical assumption of these methods is that the tracer gas is uniformly mixed in each zone of the building and remains so during the period of measurements. The major difficulty in using the methods is usually the attainment of a state of uniform mixing. The greatest error resulting from the application of the techniques to ventilation measurements is the inability to ensure the existence of this condition by the person performing the test. However, in most practical situations, an experienced professional can achieve an adequately mixed space.

The Tracer Decay Method.
The simplest and most widely used tracer gas technique for measuring ventilation is the decay method in which a quantity of tracer is injected into the building, allowed to mix and then the decay of the tracer concentration with time is measured. The ventilation rate is in this case given by the equation

$$I = \frac{1}{t} \ln\left(\frac{C_o}{C_t}\right) \tag{1}$$

where t is the time, C_o and C_t are the initial and final tracer concentrations respectively and I is the building air change rate. The air change or air exchange rate is the volume normalized ventilation or air leakage flow rate ($I = Q_L/V$, where Q_L is the ventilation or air leakage flow rate and V is the volume of the building or space being measured). This technique requires only the measurement of relative tracer gas concentrations after which the calculation required to determine I is straightforward. The measuring equipment can be located within the space or building. Alternately, building air samples containing tracer may be collected in suitable containers and analyzed off-site. Automated systems have been developed which can control the injection and sampling of tracer in multizone buildings and do real time analysis of the data. Figure 1 illustrates a schematic of the steps to be taken to perform an air change rate measurement using the tracer decay method. These steps are:

1. Inject tracer into the building

2. Wait for the tracer to mix

3. Collect at several samples at each location for a period of about two hours recording the time of each sample

4. Determine the concentration of each sample

5. Calculate the air change rate using equation 1.

From an industrial hygiene perspective, the significance of knowing I can be seen by recasting equation (1) into the following

$$C = C_0 \exp(-I \cdot t)$$ (2)

This form of the equation implies that within a room with an air change rate of 1, after one hour, an initial concentration will be diluted to 37% (e^{-1}) of its original value. In this example, if the air change rate were doubled or one waited two hours, the initial concentration would be diluted to 9% (e^{-2}) of the original value. Thus, knowledge of the actual air change rate allows an estimate to be made of the time which must elapse after a release of gas or vapor in an area before concentrations within the area are at a safe level.

Constant Injection Method

The second most common method employed to measure building ventilation rates is the constant injection method. In this method, tracer gas is injected into a test volume at a constant injection rate f while the resulting concentration is allowed to reach an equilibrium value C_∞. The ventilation or airflow rate into the test volume is determined from the relationship

$$Q = \frac{f}{C_\infty}$$ (3)

where Q is the airflow rate. Figure 3 shows an application of this method to the measurement of airflow in a duct. Note that this method provides a measure of the actual flow rate and not the air change rate. It does require that the measuring equipment be properly calibrated. The use of this method along with equation (3) requires that equilibrium be reached (under many situations this may take several hours for whole building measurements), and that the flows which are being measured remain constant over this period. These assumptions can be relaxed if more sophisticated data analysis and data collection methods are used. However, there are many situations under which the constant injection method has advantages over the decay method: in spaces which have high air change rates or in spaces in which the flow is directional and therefore it would be difficult to maintain the uniform mixing required in a decay test.

The results of a constant injection tracer test can be used directly to estimate the equilibrium level of a inert pollutant released by a constant source strength σ into the test volume

$$C_{pollutant} = \frac{\sigma}{Q}$$ (4)

where $C_{pollutant}$ is the resulting pollutant concentration.

Constant Concentration Method

The third method used for measuring building air infiltration rates is the constant concentration method. This method requires sophisticated computerized control algorithms. It is based on the principle that if the concentration of a tracer is maintained constant in a space while measuring the injection rate of tracer gas into this space required to maintain the constant concentration, then the flow of air into the space is given by the equation

$$Q_{in}(t) = \frac{f(t)}{C_{target}} \tag{5}$$

where $Q_{in}(t)$ is the airflow rate into the space from the exterior, C_{target} is the controlled level of tracer and $f(t)$ is flow rate of tracer required to maintain the target concentration at the time t. This method has the advantage of being able to measure the amount of air entering a specific zone of a building from the outside.

2.2 Instrumentation Required

Figure 2 outlines the instrumentation required to perform a tracer gas test of a building ventilation system. The basic instrumentation and materials required to perform tracer gas evaluation of ventilation systems are 1.) suitable quantities of tracer gases, 2.) a tracer gas concentration measuring device, 3.) air sampling and sample collection system, 4.)tracer gas injection systems and 5.) a data acquisition and control system. The sophistication of the system depends on the application. It can be as simple as series of air sampling containers and syringes with all analysis done in a laboratory off-site or as complicated as automated micro-processor controlled injection, multi-point sampling, data analysis and recording system used to generated data unattended over long periods of time.

In general, the evaluation of building ventilation systems requires the measurement of tracer concentrations over time at several locations in the building. This requires an air sampling system or network. The complexity of this sampling network can vary from simple manual collection systems using syringes or air sample bags to complex permanently or semi-permanently installed network of tubing, sampling manifolds and sample pumps which bring air samples from remote locations to a central point. In mechanically ventilated buildings, the building air handling system can be used as a sampling network. The decision on which type of system to use depends on the goals, resources and accuracy required by the investigation and the complexity of building being evaluated. If one requires only a one-time evaluation of a system, manual collection of data is usually the most cost effective, though in complex facilities this procedure is labor intensive.

A method for the injection of tracer into a building is also required. In mechanically ventilated buildings the injection is usually accomplished using the building air handling systems. The tracer is injected into each air supply system. In complex, compartmentalized buildings individual injections into each zone of the building is often the only method which can be used for injection of tracer. It should be emphasized that the injection of tracer into the building must be planned carefully if uniform initial concentrations are to be obtained in a reasonable length of time. This is often overlooked in the technical literature where emphasis is usually given to mixing devices such as circulating fans. The steps in planning an injection strategy for an investigation are: calculate the volume of each separate zone of the building, determine which zones are served by which air handling systems, determine the target concentration desired and calculate the quantity of tracer needed to reach the target concentration. In manual injection, it is good practice to use partial injections of the tracer in many locations in the volume being studied in order to obtain uniform mixing quickly. When using the building ventilation system, inject the tracer over a period of at least 5 minutes unless one is performing pulse tests. Certain safety considerations should also be considered. The indoor air quality concentrations should not be exceeded for the tracer used. This means that one should keep the level of a tracer to less the 1/10th its TLV and it is good practice to keep it below 1/100th its TLV. If automated injection systems are used, a safe design requires that 1/10th the TLV not be exceeded even if all tracer in the container is released at once.

2.3 Measurements in Large Buildings

This section will discuss the application of tracer gas methods to the measurement of ventilation rates in large buildings. There are two general categories of large buildings: highly compartmentalized buildings with many individual zones each served by individual ventilation equipment and buildings, though large, that are served by only a few air handlers. Figure 3 shows a schematic of an elementary school (highly simplified, the actual school had over 30 individual rooms). Each classroom as well as the cafeteria, office, gym, music room and hall were served by a separate ventilation system. The purpose of the test was to determine if the minimum setting of the ventilation systems provide sufficient outdoor air for the students. The only practical way to test this building was to use manual injection and sampling. The procedure which the authors used was to calculate the volume of each "zone of the building" and then use this volume to calculate the quantity of tracer which should be injected into each zone to produce an initial concentration of 10 ppb - at the upper end of the linear range of the monitor used. The determined quantity of gas was injected by the authors and a custodian of the school over a period of a few minutes. A period of one half hour was allowed for the tracer gas to mix in the space. For the next two hours the tracer decay was monitored by collecting 60cc syringe samples in each room. The results of this test are shown in Figure 4. It was planned to collect 4 samples per decay; however the collection of samples was interrupted for a period of about hour due to changing of custodians and only three samples were collected in some rooms. With four data points, there was no problem accurately determining the air change rate in a room. With three data points, if they did not lie on a straight line as for classroom 105, a large error or uncertainty occurred in the measurement (the numbers in parentheses in Figure 4 are the errors in the air change rate). For the gym which also only had three data points, they all lay on a straight line and therefore an accurate measurement could be made.

Figure 5 shows a schematic of a typical office building HVAC system in which a few large air handlers provide air to large zones of the building. In such a building the air ducts can be used for the injection and sampling of tracer in the building. In such a system, tracer is injected into the supply air and sampling is done in the return air. Figure 5 shows typical injection and sampling point for such a test. Figure 6 showed the tracer concentrations in the space measured with an automated tracer monitoring system. The economizer cycle is in operation during the test. The ventilation rate during the day varied from approximately 1.3 to 2.0 air changes per hour. The air change of rate when the fans were off was approximately 0.3 changes for the previous night and 0.13 the next night.

It should be noted that the ventilation and infiltration rates can be highly weather dependent. The building ventilation and air infiltration rates for the above office building are shown in Figure 7 as a function of inside-outside temperature difference. Building ventilation rates (open diamonds) are between 0.4 to 2.2 air changes per hour during periods when the building is occupied with the most typical values being about 1.0 to 1.2 air changes per hour. The extreme of 0.4 occurs during extremely hot summer conditions or extremely cold winter conditions. For comparison, the new ASHRAE ventilation standard requires 20 cfm per person for office buildings. This is equivalent to 0.8 to 0.9 air changes per hour if the building is occupied at a density of one person per 135 ft.^{2} and if the 20cfm requirement is to be satisfied.

The building air leakage (uncontrolled air exchange when the HVAC fans are off during unoccupied hours - solid squares) is between 0.2 and 0.4 air changes per hour. This is a building designed to be energy efficient and these values indicate that the exterior walls of the building are not tight by what we would consider typical of US office buildings. (Note: The surface to volume ratio of an office building is about 1/6 th that of a home. Therefore the walls of this building are equivalent to the walls of a house with an air leakage of about 1.2 to 2.4 air changes per hour - very loose).

A detailed examination of the data shows that the air handling system can easily control the amount of air required for the building. The outside air is well distributed and there is little or no evidence of short circuiting of the outside supply air or poor mixing due to the operation of the variable volume air handling system. In the summer and warmer periods of the fall and spring the typical operational mode of this air handling system runs the system at 100 percent outside air from early morning to a point in the day when the outside temperature reaches about 26 degrees C at which point the system is run at between 10 to 20 percent outside air. In the winter an economizer mode of operation is used in which the amount of outside air was determined by the cooling requirements of the building.

Figure 8 summarizes the results of the weather dependence of the building ventilation rate in eight federal office buildings. With the exception of the Federal Building in Springfield, MA , these buildings exhibit the phenomenon of low ventilation in hot summer conditions (the Anchorage building never reached those conditions) and cold winter conditions. In buildings with leaky exterior envelopes, in very extreme winter conditions, the exchange rate is dominated by natural infiltration and will increase with decreasing exterior temperature (see the paper by Grot and Persily [1985]).

2.4 Measurement of Airflows in Ventilation Systems

Tracer gas measurements can also be used to measure and characterize airflows in ventilation systems. The details of the procedure used depends on the building ventilation system. Figure 9 shows a schematic of a commonly encountered office ventilation system with recirculation. Since much of the building air is recirculated, typical air balancing procedures cannot be used to determine the amount of outside air supplied to the space. This can be done with a tracer gas using the procedure shown in Figure 10. Tracer is injected at a constant flow rate into the supply duct of the building. Tracer concentrations are measured upstream and downstream of the injection point in the supply duct, in the space of the building and in the return air. Using mass balance equations, one can determine the air supply flow rate, the percent of outside air supplied and the percent of building air infiltration. This method is more accurate then those typically used by air balancing technicians. The amount of exterior air reaching a zone can also be determined by measuring the concentration increase caused by an injection of a known amount of tracer in the building supply air.

2.5 Ventilation Effectiveness

There has been great interest in the ventilation research community over the concept of ventilation effectiveness and ASHRAE currently has a standards committee which is developing tracer gas test methods for assessing ventilation system effectiveness. The industrial hygienist should be aware that there is still considerable technical discussion of the concept of ventilation effectiveness and how it is to be measured. If one reviews the technical literature on the subject, there are probably over twenty different definitions of ventilation effectiveness. There are however two which seem to be used more frequency

than others: the concept of concentration or extraction efficiency and the local age of air definition of ventilation effectiveness. To measure the concentration or extraction efficiency, a constant injection test is performed with the tracer source in the occupied space. When equilibrium is obtained, the tracer concentrations are measured in the occupied space and in the exhaust air. The extraction or concentration efficiency ε_j at a location j is defined as

$$\varepsilon_j = \frac{C_{exhaust}}{C_j} \tag{6}$$

where $C_{exhaust}$ is the exhaust air tracer concentration and C_j is the tracer concentration at a location j in the space.

The age of air definition of ventilation effectiveness attempts to determine how long a package of air remains in a space. This is accomplished by creating an uniform concentration of tracer gas in the space by using a constant injection of tracer into the building supply air. The injection of tracer is turned off and the average concentration of the tracer during the decay at a given location is measured using a integral sampling technique. The local mean age of air is defined as

$$\tau_j = \frac{\int C_j(t)dt}{C_j(0)} \tag{7}$$

where $C_j(0)$ is the initial tracer concentration. The local ventilation efficiency is defined as

$$e_j = \frac{\tau_n}{\tau_j} \tag{8}$$

where τ_n is the inverse of the whole building air change rate.

2.6 Ventilation Performance from CO_2 Measurements

Many industrial hygienists have been using CO_2 measurements to estimate building ventilation rates. A detailed check of this procedure has been undertaken by the National Institute of Standards and Technology (Persily and Dols [1990]). They found that this technique is not reliable when the building is occupied. During periods when the building is unoccupied, the decay of CO_2 will yield an estimate of the building ventilation rate if sufficient CO_2 remains in the building. Figure 11 shows a typical weekly and daily profile of CO_2 levels in the Portland Federal Building. Note that steady state is never obtained. This non-attainment of equilibrium leads to a large over estimate of the ventilation rate occurring in the building.

Figure 12 shows the maximum daily CO_2 level as a function of air exchange rate. Carbon dioxide levels are seldom over 600 ppm on a building average and only a couple of times ever over 1000 ppm at any location in the building. (The new ASHRAE standard proposes a maximum level of 1000 ppm; complaints from building occupants begin to occur when levels exceed 600 ppm). A closer examination of the hourly CO_2 levels in the building shows that the measured levels are never at steady state and usually have two daily peaks - one around 11 am and the other around 3 pm. This is also shown by the values of the source strength constant of the fitted curve in Figure 12. The value of

source strength constant, 100 ppm per hour, is approximately 1/3 the value of the source strength constant derived from equilibrium theory based on the occupancy of the building.

Carbon dioxide levels can be used to indicate that a ventilation systems is not performing properly; however they cannot be used to give a accurate value for the building ventilation rate.

2.7 The Estimation of Contaminant Source Strengths

It is possible to use tracer gas measurements along with pollutant measurements to estimate contaminant source strengths. The contaminant source strength can be defined as

$$\sigma_{pollutant} = \frac{\Delta C_{pollutant}}{Q_{in}} \tag{9}$$

where $\Delta C_{pollutant}$ is the difference between the interior and exterior pollutant concentrations and Q_{in} is the total inflow rate measured by a tracer test for the space.

Figure 13 shows the effect of building ventilation rate on the total VOC concentration. Table 4 gives the effect of ventilation rates on the individual compounds detected in the building. The measurements were made on four different occasions over a period of 17 months (see Grot et al., 1987 and Hodgson et al., 1990). On each of these dates the building was being operated with four distinct air exchange rates (0.5, 1.36, 0.24 and 1.99 changes per hour) due to the prevailing exterior weather conditions. The curve in Figure 13 represents the predicted level of VOC's in the building using the source strengths estimated from the VOC levels and the air change rates. The source strength of total VOC was remarkably constant over the 17 month period between the first and last measurements. We have measured and identified 37 volatile organic compounds in the interior building space. These are given in Table 4. There were 5 oxygenated compounds, 6 halogenated compounds, 16 alkanes, 6 cycloalkanes and alkenes, and 5 aromatic hydrocarbons found in the building. All are at levels less than 1/1000 th of the OSHA standard environmental levels of industrial work spaces. (Note: The ASHRAE standard recommends that for indoor air quality the level be not more than 1/10 of OSHA SEL's.) The largest amount of the mass of the VOC's is concentrated in the alkane class (C_{10} to C_{12} branched decanes and undecanes). These are not particularly irritating compounds and there are no OSHA recommended levels for these substances. However very limited studies done in Denmark by Molhave [1983] indicated that many complaints will occur when the total levels of VOC's exceeds 5 mg/m^3 and it has been recommended by researchers (Tucker, 1988) at EPA Research Triangle Park that a prudent target level for total VOC's be 1 mg/m^3. All four measurements sets that were made in the building were greater than 1 mg/m^3 and the building exceeded 5 mg/m^3 when the ventilation rate was below 0.5 air changes per hour. Though operating the ventilation system at 100 percent outside air would keep the levels near the target of 1 mg/m^3, identification and limitation of the sources is a better strategy.

Figure 14 shows the short term variation in VOC source strengths over a period of four days, beginning at 6 am on Friday before the building was occupied, carrying over the weekend when only a few occupants were in the building and continuing till 6pm on Monday. It is clear from the data in Figure 14 that the source strengths were greatest during the occupied day and low in the evenings even when the HVAC was off and on the weekends when few people were in the building. Therefore the source of VOC's was

activity related and closer comparison of the chromatographs of samples in the building and those of the liquids used by the copiers in the building confirmed that the liquid copiers were the major source of VOC's.

2.8 Multi-Tracer Techniques

The above tracer gas measurement techniques generally employ a single zone model of a building. In some cases, a single zone is not adequate to model a building's infiltration characteristics, or one is interested in the airflows between various zones of a building. In these cases, multi-chamber building models and multiple tracer gas measurement techniques are used. The use of multiple tracer techniques for building airflow studies began in the middle seventies. A table of tracer gases that have been used in ventilation measurement applications has been provided in table 3. Multiple tracer measurements often involve the use of gas chromatographs designed to determine simultaneously the concentration of the different gases; however, separate continuous infrared analyzers have also been successfully used for simultaneous analysis of SF_6, CO_2 and N_2O (Perera et al. 1983).

An illustration of the simultaneous chromatographic separation of six refrigerants is provided in Figure 15. Note that the time for a single measurement of the six gases is on the order of ten minutes. A throughput time of this magnitude may preclude the use of a single gas chromatograph in a constant concentration measurement. If one desires analysis times on the order of minutes, it is necessary to have several gas chromatographs available.

Multiple tracer gas measuring systems have been developed using both decay and constant injection techniques. In decay measurements, a tracer or several tracers are released at various locations as pulses (see Axley and Persily, 1990), and their concentrations are monitored in the various zones over time. Several measurement systems employing the decay method have been developed, including those of Prior et al. (1983) and Irwin et al. (1984). Both systems employ electron capture detectors, to measure refrigerants in the first system and perfluorocarbon tracers in the latter. I'Anson et al. (1982) also employs a decay system involving refrigerant tracers. In Figure 16 we show representative data from their paper in which two tracers are used to study the airflow between the upper and lower levels of a building. The refrigerant $C_2Cl_2F_4$ is injected downstairs and CCl_2F_2 upstairs. The movement of tracer between the zones and the subsequent decay is evident in the data. Analysis of these data yields the airflow rates between the two zones and from each zone to the outside.

A constant injection technique has been employed by Dietz and Cole (1982) and Dietz et Al. (1984a) using several perfluorocarbon tracers and passive samplers. In these measurements a different tracer is released at a constant rate into each zone and the average concentration in all zones is determined over the measurement period. From the injection rates and the average concentrations, one determines the airflows of interest. An example of this technique was given in the first article. This technique is used for long term studies of air infiltration behavior, consequently it is not generally useful in industrial hygiene applications.

There have been other multiple tracer studies including those of Foord and Lidwell (1973 and 1975) studying ventilation rates and interzonal airflows in hospitals. Lagus (1977) suggested the use (shown in Figure 17) of multiple tracer gases in multistory buildings for evaluating simultaneously infiltration rates on individual floors and airflow rates between floors by injecting a different tracer on each of three adjacent floors. Figure 18 shows an example of a constant injection measurement of ventilation and interzonal airflows in two adjacent, negative pressure laboratory rooms (Lagus 1984). The two laboratories are

each under negative pressure, and a different tracer gas is injected into each laboratory at a constant rate. Using the equilibrium concentration values, along with the known injection rates for the tracers, it is possible to calculate ventilation rates using equation (I-9) given in the appendix. The equilibrium tracer concentrations are shown in Figure 19. The measured airflow rates agree well with the ventilation rates measured by tracer decay. In addition, note the flow of the refrigerant $CBrF_3$ into the lower laboratory. Since the ventilation rate is now known on the lower floor, the steady value of $CBrF_3$ concentration in the lower floor, coupled with the measured value of the concentration in the upper floor, allows one to calculate leakage flow from the upper floor to the lower floor. This evidence of undesired airflow between the laboratories led to the laboratory owner taking remedial action. Similar analyses can be extended to additional floors or zones using additional tracer gases. However, the experimental complexity of these measurements usually requires the supervision of an experienced investigator. Nevertheless, in those cases where multiple flow regimes need to be analyzed simultaneously, a multiple tracer technique is perhaps the only valid experimental approach. A detailed description of this method is beyond the scope of this article. Industrial hygienists should be aware that these interzonal airflow measurements can be made if required. However, in most situations, methods described below will provide more direct answers to contaminant transport or containment questions.

3 Airborne Hazardous Substance Assessment By Tracer Gas Methods

This section will present a description of several applications of tracer gas analysis to evaluate the performance of ventilation systems used for hazardous substance or contaminant containment and to investigate airflow patterns induced by these systems. Within this general area, tracer gas methods can be used to:

- Evaluate the performance of secondary ventilation systems used for hazardous containment.

- Delineate airflow patterns to allow proper placement of health and safety monitors.

- Document the existence of and provide quantitative estimates of potential hazardous gas or vapor leakage from one area to another.

- Evaluate areas within buildings for suitability as temporary safe havens.

- Document the existence and magnitude of exhaust gas re-circulation and re-entrainment within occupied buildings.

Each of these topics will be discussed below.

3.1 Fume Hood Re-Entrainment and Re-circulation Testing

Fume hood re-circulation studies are normally performed by injecting a known flow rate of tracer into a fume hood and then monitoring the room surrounding the fume hood for the presence or absence of tracer gas. Injection into the fume hood is performed by using a cylinder of diluted compressed tracer gas as a source. Figure 20 provides a schematic drawing of the tracer source cylinder within a fume hood. The flow rate of tracer is controlled by an orifice or an electronic mass flow controller. The fume hood duct concentration is

$$C_{duct} = \frac{C_{tracer} \times Q}{F_D} \qquad (10)$$

where

Q = Tracer injection rate
C_{duct} = Fume hood concentration
C_{tracer} = tracer injection concentration
F_D = Duct flow rate

To check for re-circulation, samples of laboratory air are obtained at a number of locations over a limited time period and are analyzed for the presence or absence of tracer. If a concentration of tracer, C_{lab}, is measured within the laboratory, this implies re-entrainment or re-circulation of fume hood exhaust. The ratio of the laboratory concentration to the duct concentration yields a measure of the dilution of the stack effluent

$$D = \frac{C_{lab}}{C_{duct}}$$

(11)

where D is the dilution ratio.

Knowledge of this dilution ratio allows assessment of the potential for hazard to laboratory personnel from a particular species being exhausted through the fume hood. This ratio is applicable to all non-reacting species. Thus a dilution ratio measured by the tracer technique can be used to estimate concentrations of hazardous gases and vapors which may be released in the fume hood being studied. The results of the test can be used to adjust the fume hood exhaust to an optimal level, ensuring the safety of the workers at a minimum expenditure of energy.

The evaluation of fume hood containment using a tracer gas was pioneered by Caplan and Knutson. The American Society of Heating, Refrigerating and Air-Conditioning Engineerings (ASHRAE) promulgated the standard ASHRAE 110 for evaluating fume hoods using the methods developed by Caplan and Knutson. In this test procedure, a mannikin is placed in front of the fume hood and sampling for the tracer is done in the region near the mannikin's face.

This technique has led to an increased understanding of the influence of the user and the local ventilation system on the performance of fume hoods. Unfortunately, this technique only addresses the problem of the adequacy of the fume hood to contain flow within it.

There is a second and potentially more important question which can be also addressed with a tracer technique, namely how much, if any, of the effluent being contained and exhausted by the fume hood is making its way back into the laboratory, laboratory building, or surrounding buildings?

The return of exhausted gas into the building or laboratory is known as re-circulation or re-entrainment. While the words are often used interchangeably, in this article, re-circulation will be taken to mean exhaust gas which enters the building (or laboratory) before it is expelled to the outside air. Re-entrainment will be taken to mean exhaust gas which has been expelled from the building through a vent or duct and subsequently reenters the building due to wind induced airflow patterns around the building.

A simple constant flow tracer test can be used to investigate and quantify potential re-circulation and re-entrainment of fume hood exhaust. Tracer is injected into the fume hood and the surrounding area is sampled for the presence of tracer gas. Injection can be either continuous or pulse as described in the first article. Sampling can be effected by either grab samples or time averaged bag samples.

Table 5 presents the results of tracer re-circulation tests performed for a research laboratory building. Data are presented as dilution ratios as described in the first article. A total of 32 fume hoods were evaluated. Of this total, 7 hoods evidenced measurable re-circulation. In order to eliminate the possibility of back streaming of tracer from the hood into the laboratory, tracer was injected directly into the exhaust slot at the top of the flume hood enclosure. The measured dilution factors ranged from roughly 700 to 7000. Note that re-circulation was measured not only in the rooms containing the hoods, but in 4 cases, measurable tracer was found in adjacent rooms. All of the hoods were of an older design in which a booster fan is located in the ductwork immediately above the false ceiling in the laboratory. The run of duct immediately downstream of this booster is therefore under positive pressure and is the most likely source of exhaust re-circulation. From these measured data one can calculate the potential hazard for any assumed release within the fume hood. For instance, for several of the hoods tested a release of bromine into the hood sufficient to result in an exhaust duct concentration of 1000 ppm would result in the re-circulation of approximately 1ppm of bromine into the lab rooms. This concentration could present a health hazard and as such would require remedial action.

3.2 Evaluation of Exhaust Re-Entrainment and Re-circulation

Figures 21 and 22 show two examples of exhaust re-entrainment and re-circulation in a building. Figure 21 shows the re-entrainment from an exterior contaminant source, while Figure 22 illustrates re-circulation caused by the re-entrainment of a building exhaust. Both situations can be tested using tracer methods which will provide quantitative information to assist in evaluating the potential hazard due to re-entrainment. There are two methods which can be used to evaluate this re-entrainment: the steady state method which consists of injecting tracer gas at a constant flow rate in the vicinity of the source being evaluated and the pulse technique in which a specific quantity of tracer gas is injected in a manner consistent with an anticipated occurrence of the spill or exhaust of contaminant at the source. The first method yields a worst case evaluation of re-entrainment; the second more closely mimics a spill or inadvertent release and yields an estimate of the resulting dosage to which a person would be subjected. In the steady state method, the concentration at various parts of the building and in various air intake ducts is measured. Several useful measures can be derived from these tests. In the case of an air intake duct, the percent of exhaust re-entrained can be calculated from the equation

$$\% = \frac{F_D \times C_D}{Q_s} \tag{12}$$

where
    F_D is the duct flow rate
    C_D is the tracer concentration in the duct
    Q_s is the source flow rate of tracer

In many buildings, the air intakes are not the only location of re-entrainment. Areas of the building under negative pressure such as return air plenums, elevator shafts, entrance ways and points of air infiltration can also cause contaminant re-entry into the building.

The second method of evaluating the potential for re-entrainment is to inject a pulse of tracer and measure the integral of the concentration over the period of time for which exposure is to be evaluated. This can be done either by using a continuous monitor at each location or by employing an integrated sampling method such as the filling of a sample bag with a constant flow pump at a given location. The specific dosage of tracer at a given location in the building is determined by

$$d = \frac{C_{bag} \times t}{m_{tr}} \tag{13}$$

where
> d is the dosage
> C_{bag} is the concentration in the bag
> t is the length of the time interval
> m_{tr} is the mass of tracer injected

The dosage caused by any spill of a hazardous substance of mass M_h is then given by

$$dosage = d \times M_h \tag{14}$$

One drawback to the pulse test is that the result of a single measurement is much more dependent on local wind conditions than for a steady state test.

Table 6 gives the results of a re-entrainment test performed in a school in such that were indoor air quality problems. Each classroom of the school was serviced by a combination ventilation and heating/cooling unit (see Figure 26.). The flue for the gas heater in the HVAC unit was located on the opposite side from the air intake. The flue gas was exhausted by a power blower downward toward the roof surface. Several of these units were located on the roof of the school in close proximity to each other and at times under a parapet with the exhausts and intakes of adjacent units near each other. The data in Table 6 show that about 0.1 to 0.2 % re-entrainment of the flue gas through the air intakes of units adjacent to the fired gas heater in unit C. After a half an hour, the concentration of tracer in each classroom resulting from flue re-entrainment was measured. If these data are used in a mass balance model, the amount of tracer entering each room is about five times greater than the quantity entering through the air intake (0.8% to 1.6% of the quantity of tracer released). There seems to be a hidden leakage under the unit into the return air plenum serving the classrooms which is being re-circulated to the interior of the classrooms.

An estimate of the level of pollutants in the classrooms generated by the gas heater due to re-entrainment of the flue gases can be derived using the data from re-entrainment tests and data from ventilation tests by considering typical emissions from a convective gas heater. The steady state level of the pollutant in the room can be determined from the equation:

$$C = \frac{G \cdot D}{ACH \cdot Vol} \tag{15}$$

where:
> C is the pollutant concentration in mg/m^3
> G is the pollutant generation rate in mg/hr
> D is the measured dilution ratio
> ACH is the air change rate in air changes per hour
> Vol is the volume of the room in m^3

The worst case of re-entrainment in this school, 4.4%, was measured for classroom K. Table 7 gives the results of a typical use of measured dilution ratios and room air change rates to estimate the levels of CO_2, CO and NO_x which could result from the enter of flue gas into the classrooms.

3.3 Hazardous Substance Containment Testing

Tracer gas methods can be used to test the integrity of a hazardous substance containment system or a secondary containment ventilation system. Figure 23 shows a schematic of a test procedure for implementing a secondary containment ventilation system test using a tracer gas. Monitoring is implemented by injecting a constant flow of tracer into a hazardous containment area and monitoring the periphery for the presence or absence of the tracer. If no tracer is found within the non-hazardous periphery, no breach of containment has occurred. If tracer is detected within the non-hazardous area of the facility, this presents unambiguous evidence of a containment breach. The magnitude of the tracer concentration yields quantitative information on the magnitude of the integrity breach. If the ventilation rate of the non-hazardous area is known or measured, then an effective leakage rate of the breach can be calculated. The effective leakage rate can be defined as the fraction of total leakage from a containment area into a non-containment area. The effective leakage rate is given by

$$L_{eff} = \frac{C_p}{C_{con}} \times I_p \tag{16}$$

where
L_{eff} is the effective leakage rate
C_p is the tracer concentration in the non-hazardous area
C_{con} is the tracer concentration in the containment area
I_p is the ventilation flow rate in the non-hazardous area

If it is desired to know the probable concentration of a given airborne contaminant at a number of locations which are separated from the containment area, but for which there exist potential flow paths, tracer concentration measurements can be made at these locations and a dilution ratio calculated

$$D_{loc} = \frac{C_{loc}}{C_{con}} \tag{17}$$

where
D_{loc} is the dilution ratio
C_{loc} is the tracer concentration at the location of interest
C_{con} is the tracer concentration in the containment area

The dilution ratio at a given location can be used to assess the hazard to persons working at that location if the concentration of the hazardous substance in the containment area is known.

Figure 24 depicts the use of tracer to test for the leakage from a hazardous area into a control room. Release of a tracer at a constant flow rate in the hazardous area, coupled with measurements within the controlled area, can be used to evaluate the potential hazard to persons working in the control room and to assess the ability of the containment system to protect the control room personnel.

Figure 25 suggests a method for measuring and locating leaks in a fresh air supply duct to a control room. In many HVAC systems, especially in return air and fresh air supply ducts which are under negative pressure with respect to areas through which they pass, inadvertent and non-design leakage can be a problem if these ducts pass through hazardous areas. A constant injection of tracer into the duct, followed by subsequent measurements along the duct, affords both the measurement of the flow rate through the duct and the possibility of locating the major leakage points.

In many cases a room, such as a control room or a computer room in a process plant, is isolated in a ventilation sense from the remainder of the plant. Since duct work is seldom truly leak tight, it is possible for inleakage into the return portion of the control room ductwork to engender measurable concentrations of hazardous gases or vapors within the control room.

A tracer test can be used to measure the amount of unwanted inleakage into the control room. Knowledge of this inleakage, as well as any potential hazardous concentration in the area of the return ducting, allows calculation of actual hazardous concentrations (and hence exposures) within the control room.

A typical scenario is presented in figure 23. A uniform tracer concentration is established in the volume surrounding the return duct, and any resulting tracer concentration in the control room is measured. A schematic of the actual ventilation flows in a typical control room is presented in figure 27. Note that the makeup air is provided from outside the HVAC Equipment Room. For the test shown in figure 23, there was concern about potential contaminants within the HVAC Equipment Room migrating to the Control Room via return duct inleakage. A uniform tracer concentration was established within the HVAC Equipment Room and the resulting tracer concentration within the Control Room was measured. The fact that ANY tracer was measured in the Control Room established that Control Room inleakage via the return duct could result in contamination of the Control Room.

A simple mass balance, keyed to figure 27, yields the equation

$$Q_{in} = \frac{(C_s \cdot Q_s - C_r \cdot Q_r)}{C_{hvac}}$$

(18)

where

Q_{in} = Control Room inleakage rate

Q_s = Flow rate in Control Room Supply Duct

Q_r = Flow rate in Control Room Return Duct

C_s = Tracer Concentration in Control Room Supply Duct

C_r = Tracer Concentration in Control Room Return Duct

C_{hvac} = Tracer Concentration in HVAC Equipment Room

Measured data for this test are provided in Table 8. The inleakage shown was for a run of duct approximately 100 feet long. Though this example was for a control room, it could easily apply to inleakage of corrosive compounds into a control computer room in a chemical process plant.

3.4 Airflow Pattern Testing/Hazardous Substance Migration

Perhaps the simplest use of a tracer gas is to track the airflow patterns within a complex industrial environment. In Figure 28, we illustrate the use of a tracer to provide both qualitative and quantitative information about the flow patterns within a chemical process facility which was designed to destroy quantities of obsolete toxic material. In the facility shown, toxic materials enter through the Supply Conveyor Room. The toxic materials were utilized in small containers, a number of which were in turn stored in larger transportation casks. The material within the individual containers was in a liquid state, but its vapor was extremely toxic. Within the plant, incoming transportation casks were received and stored in the Supply Conveyor Room. The transportation casks were then transferred into a negative pressure glove box where they were opened. The smaller containers were taken out of each

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cask inside this glove box. From here the containers were conveyed to a rotary kiln for destruction. The kiln was located in an adjacent building. Health and safety monitors were located at locations B and E in Figure 28. In order to assess the potential hazard that a leaking cask in storage could pose to unprotected personnel in the Disassembly Room, a test was performed in which a constant flow of sulfur hexafluoride tracer gas was injected into the Supply Conveyor Room to simulate a leaking cask. Grab samples were taken throughout the Disassembly Room and analyzed for the presence of tracer gas. Results are provided in Table 9. Note that the tracer concentration within the Supply Conveyor Room was not greatly diluted by flow into the Disassembly Room. Furthermore, the concentration at location B was lower than practically any where else in the room, even though this was the location of one of the two health and safety monitors. Additional tracer gas measurements showed that it took approximately 4 minutes for the concentration at location G and D to become substantial while it took roughly ten minutes for the concentration to rise to a comparable level at both locations E and B. Thus by means of a simple tracer test, it was possible to quantify the hazard within the Disassembly Room due to leakage of vapor within the supply conveyor room and to evaluate the appropriateness of the location of the health and safety monitors within the facility.

In industrial facilities health and safety monitors (chemical, hazardous vapor, or radiation) are often located for the convenience of installation and maintenance personnel in the absence of more compelling data such as the tracer data presented above.

The concept of using a tracer to examine flow patterns can be expanded to encompass the investigation of multiple flow paths simultaneously. All that is required is a distinct tracer gas for each flow path to be investigated. For the purposes of the test a flow path may be an identifiable path or it may be a single air handling unit within a complex building. Practically speaking, testing with more than three or four gases simultaneously is very tedious and time consuming. The data, however, can be extremely informative.

An example in which three flow paths were tracked simultaneously using three tracer gases (Halocarbon 13B1, Halocarbon C-318, and Perfluorodimethylcyclobutane, PDCB) is provided in figure 2 (Lagus et. al, 1988). This shows one level of a seven storey auxiliary building within a plant complex. Tracer source locations are marked by large letter S's, while sampling locations are marked by X's. Source injections were performed continuously over a period of four hours during which time grab samples were taken throughout the building at distinct locations at selected timed intervals. These samples were then analyzed on-site for the presence of tracer gas.

The requirement for this testing arose due to the discovery of occasional episodes of higher than expected concentrations of hazardous gases at locations that should not experience such levels with the ventilation in its normal mode. The tracer source locations were chosen to correspond to locations which could act as sources for the gases. The ventilation system was designed to contain the hazardous gases in these areas. In Figures 29 and 30, the solid arrows denote the anticipated flow directions based on the design of the ventilation system. The dashed arrows show the actual flow directions as discerned by the tracer measurements. It is apparent that for some of the locations the ventilation system was not performing as designed. It should be emphasized that immediately prior to performing these tests, the entire building ventilation system had been balanced by a licensed test and balance (T&B) contractor, thus the flow directions actually observed are not due to the system being out of balance.

For several of the flow tests performed, tracer concentrations were measured on adjacent floors implying the existence of flow or migration between floors. Gas migration was also seen to occur up the stairwells. For some of these cases it was possible to quantify the amount of flow entering particular areas on the adjacent floor. An example of tracer data which can be used for this type of interpretation is shown in Figure 31. In one particular room of interest, an inleakage of 3 cfm was detected during this testing. Note that the transient build-up of concentration shown in figure 4 has gone to completion and that the resulting tracer concentrations are approximately steady state. Reference to the discussions surrounding equation (16) apply to these types of problems, and the mathematical analysis presented there, allows one to calculate leakage flow rates into selected rooms. This technique can be extremely useful when an estimate is required of the potential threat created by the accidental release of a hazardous gas or vapor within an area of a plant. It is important to realize that the simple interpretation described in the first article cannot be undertaken if the tracer concentrations are changing as a function of time.

Another useful assessment of the performance of this same auxiliary building ventilation system was made by placing the building ventilation system in its emergency mode. By design, in the emergency mode, the ventilation system splits the building into two zones separated at the fourth level. The idea is that in the event of a malfunction, the lower floors could become contaminated. Hence, any air in this region should not interact with air above the fourth level. It is passed to the outside through an elaborate filter system. The question naturally arose during the course of these measurements as to the degree of ventilation isolation. Accordingly, a massive tracer release was undertaken in the lower floors with subsequent sampling on the upper floors. No tracer was detected over an extended period. This lack of detectable tracer coupled with a knowledge of the tracer concentration in the lower floors could be used to place an upper bound on any potential leakage across the fourth floor boundary. Equation (16) can again be used where the concentration measured (or should we say not measured) corresponds to the detection limit for the particular tracer. For the building shown above, the ratio of tracer below and above the fourth floor was 50,000 to 1. This implies that any flow across the boundary was negligibly small.

3.5 Temporary Safe Havens

A safe haven is a room or a building that, by virtue of its specific design and construction, will afford full protection to a limited number of personnel for the expected duration of an exposure to a toxic release. Safe havens (typically control rooms required for the orderly shut down of a process) usually require special HVAC system features and/or supply clean breathing air for a designated number of personnel.

A temporary safe haven does not provide the same level of protection as a safe haven, but can provide valuable time at the onset of a crisis such as the accidental release of a hazardous or toxic gas. Like a safe haven, a temporary safe haven is a room or building that provides protection from toxic fumes, but the protection is for a limited time--possibly less than the duration of exposure to a toxic cloud.

Most toxic gas release incidents are of short duration or are controlled quickly. Therefore, occupancy of a temporary safe haven is preferable to immediate evacuation and, in most cases, affords full protection. Even for prolonged releases, a temporary safe haven provides additional time while the emergency is assessed and allows first respondents to organize an orderly evacuation if necessary. For the temporary safe haven concept to be useful in an emergency response function, it is important to know the minimum protection afforded by specific buildings so that protected assembly points can be selected. Many plant sites have hesitated to designate temporary safe havens because no quantitative method was available to ensure adequacy of candidate buildings or rooms.

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A method based on the tracer concentration decay method (ASTM Standard E741) has been suggested as a means to evaluate buildings or rooms within buildings for suitability. By first measuring the air infiltration rate as described in the first and second articles, Jann (1989) provided a series of nomographs based on the measured air exchange rate in order to estimate the safe time within a temporary safe haven for several outside toxic challenges. These are reproduced in Figure 32. Since most toxic releases tend to be of rather short duration, nomographs such as these can be used to assess the degree of protection afforded by various potential temporary safe havens.

3.6 Secondary Ventilation System Testing

In many industries, especially in the semiconductor fabrication industry, process and fabrication equipment is often equipped with a secondary ventilation system to provide extra protection to plant personnel in the event of the release of chemicals (usually gases) used in the manufacture of semiconductors. In the event of a hazardous gas leak, the effectiveness of secondary exhausted enclosures becomes critical to prevent harmful exposure conditions. Reliance on gas detection for worker protection is ineffective in the absence of effective leak containment. By the time a large leak is detected harmful exposures could have already occurred. Detector cycle times and detector sensitivity limitations can lead to a considerable interval elapsing between introduction of a hazardous gas into the work area and any resulting emergency action.

Secondary enclosure exhaust ventilation and detection controls for hazardous gases used in the semiconductor industry are commonly installed according to equipment manufacturer specifications or theoretical design criteria. Before the advent of tracer techniques it was not possible to quantitatively test the working of these systems under actual operating conditions. A typical test setup is shown in figure 8. Tracer is injected into the enclosure at a known rate and the periphery of the enclosure is sampled for the presence of tracer (Tubby, 1991). It is possible to interpret the tracer resulting concentration in terms of concentration of process gases by means of ratio analysis as explained the discussion related to equation (17).

Typical data obtained from a test of a Chemical Vapor Deposition system are presented in table 9 (Orcutt, 1988). In this table, the dilution ratio normally measured has been converted to an equivalent concentration of process gas released within the enclosure. Note that for many of the gases used in semiconductor fabrication the equivalent leakage concentrations shown in this table are significantly in excess of allowable TLV values. There is now a SEMI Standard, S2-91, which mandates this type of testing for new semiconductor fabrication equipment.

Summary

This article has provided a summary of various tracer gas techniques which can be used in industrial hygiene investigations. It has discussed the three tracer gas methods used to measured building ventilation rates and building airflow rates. The testing of fume hoods for re-entrainment and re-circulation has been described. Methods using tracer gases for assessing the release of hazardous substance in a facility were outlined. The evaluation of computer models for predicting contaminant dispersal and air or contaminant movement using tracer gas testing was touched on. In future articles, each of these topics will be expanded upon to provide the reader more complete explanation of the use of tracer gas techniques for industrial hygiene investigations.

This article has presented the various tracer gas methods which can be used to assess the performance of building ventilation systems. Accurate measurements of total building ventilation and air leakage rate can be made in even large complex buildings at moderate expenditure of effort and cost. More complicated tests can be performed to measure airflows between zones of the building, to assess ventilation effectiveness and to aid in estimating contaminant source strengths.

This article discussed the use of tracer gas methods for the assessment of airborne hazardous substance releases. It showed examples of the use of tracer gas techniques for tracing air flow paths, evaluating fume hoods, measuring contaminant inleakage into air ducts and areas protected by secondary ventilation systems and measuring flue gas reentrainment. It gave examples of the quantitative use of tracer data for assessing the exposures and dangers which can be caused by the release of hazardous substances.

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Table 1
Tracer Gases and Measurement Techniques

Technique	Gases
Thermal Conductivity Detector	Hydrogen Helium Carbon Dioxide
Electron Capture Gas Chromatograph	Sulfur Hexafluoride Halogenated Compounds Perfluorocarbons
Flame Ionization Gas Chromatograph	Ethane Methane
Infrared Absorption	Carbon Monoxide Carbon Dioxide Sulfur Hexafluoride Nitrous Oxide Ethane Methane

Table 2
Relative Tracer Gas Costs Taking Detectability into Account

Gas	Detectable Concentration (ppm)	Gas Volume Per Dollar		Maximum Measurable Volume Per Dollar	
	(ppm)	ft^3	(m^3)	ft^3	m^3
He	300	1.4	(0.13)	$4 \cdot 10^3$	$(4 \cdot 10^2)$
CO_2	1	7.0	(0.65)	$6 \cdot 10^4$	$(6 \cdot 10^5)$
N_2O	1	2.4	(0.22)	$2 \cdot 10^6$	$(2 \cdot 10^5)$
SF_6	$5 \cdot 10^{-6}$	0.13	$(1.2 \cdot 10^{-2})$	$2 \cdot 10^{10}$	$(2 \cdot 10^9)$
"	$5 \cdot 10^{-3}$	"	"	$2 \cdot 10^7$	$(2 \cdot 10^6)$
$CBrF_3$*	$5 \cdot 10^{-5}$	$3.7 \cdot 10^{-2}$	$(3.4 \cdot 10^{-3})$	$7 \cdot 10^8$	$(7 \cdot 10^7)$
PDCH**	$5 \cdot 10^{-5}$	$3.0 \cdot 10^{-3}$	$(2.8 \cdot 10^{-4})$	$6 \cdot 10^8$	$(6 \cdot 10^7)$

* bromotrifluoromethane (Freon 13B1)
** perfluorodimethylcyclohexane

Table 3. Electronegative Tracer Gases

Name	Symbol	Trade Name
Dibromodifluoromethane	CF_2Br_2	Freon 12B2
Trichlorofluoromethane	$CFCL_3$	Freon 11
1,1,1,-Ttrichlorotrifluoroethane	$C_2CL_3F_3$	Freon 113
Bromotrifluoromethane	CF_3Br	Freon 13B1
Qctafluorocyclobutane	C_4F_8	Freon C-318
Dichlorodifluoromethane	CCl_2F_2	Freon 12
1,2-Dichlorotetrafluoroethane	$C_2Cl_2F_4$	Freon 114
Chlorodifluoromethane	$CHClF_2$	Freon 22
Chloropentafluoroethane	$CClF_2CF_3$	Freon 115
Bromochlorodifluoromethane	$CBrClF_2$	Freon 12B1
Suflur Hexafluoride	SF_6	
Perfluorodimethylcyclohexane		PDCH
Perfluoromethylcyclohexane		PMCH
Perfluoromethylocyclopropane		PMCP
Perfluorodimethylcyclobutane		PDCB

Table 4.
Long-term variations in the concentrations and specific source strengths of individual VOC.

	CONCENTRATION (μg/m3)				SPECIFIC SOURCE STRENGTH (μg/m3-h)			
Date	8/4/87	10/14/87	1/13/88	10/28/88	8/4/87	10/14/87	1/13/88	10/28/88
Time	20:00	17-19:00	15-17:00	15-17:00	20:00	17-19:00	15-17:00	15-17:00
Vent. Rate	0.5	1.36	0.24	1.99	0.5	1.36	0.24	1.99
COMPOUND								
Oxygenated								
2-Propanol	14.8	20.2	137.2	26.5	5.8	21.9	31.6	52.6
2-Propanone	50.1	28.8	66.6	32.4	22.1	27.0	14.9	49.7
2-Butanone	40.9	6.2	15.3	5.7	19.0	2.5	2.0	7.6
Chlorinated								
Dichloromethane	32.4	2.6	13.4	2.7	15.9	1.3	2.7	5.4
1,1,1-Trichloroethane	13.5	13.8	119.7	17.1	5.4	13.6	27.5	27.0
Trichloroethene	16.4	7.2	58.2	14.8	8.2	9.7	11.0	27.6
Alkane + Cycloalkane								
2-Methylbutane	31.9	53.8	81.6	26.2	13.1	31.7	16.1	44.7
n-Hexane	11.3	10.0	24.0	9.2	5.7	6.7	3.7	14.6
Cyclohexane	5.7			2.7	2.4			4.2
n-Heptane	4.8	3.1	12.6	3.6	2.0	2.7	0.2	5.5
3-Methylhexane	6.0	4.0	14.7	3.5	2.4	3.1	0.0	5.2
Methylcyclohexane	5.1			1.7	2.4			2.5
2,2,4-Trimethylpen-tane	2.4	1.8	8.0	3.0	1.0	1.3	0.7	6.0
1,4-Dimethylcyclohex ane	3.1				1.6			
n-Nonane	39.6	10.6	149.1	33.9	19.7	11.4	35.3	63.0
2,2,5-Trimethylhexane	2.4				1.2			
Isopar 2	147.0	82.5	638.7	95.4	72.8	104.2	151.7	179.7
n-Undecane	115.6	57.3	831.3	48.3	55.2	71.3	196.8	85.7
n-Dodecane	49.1	10.6	280.8	10.9	21.8	5.9	67.0	17.6
n-Tridecane		6.0	111.9	8.5		5.8	26.2	13.8
n-Tetradecane		36.1	245.3	27.0		43.0	57.9	49.5
Aromatic								
Toluene	60.4	81.3	91.0	33.1	22.7	80.9	13.7	50.2
Ethylbenzene	11.8	7.0	18.7	7.5	5.3	4.9	2.3	11.4
1,2-Dimethylbenzene	17.2	8.7	25.8	8.1	7.6	5.8	4.1	12.0
1,3-,1,4-Dimethylbenz ene		18.1	54.5	18.3		11.7	8.8	26.6
1,3,5-Trimethylben-zene	4.1				1.6			
TOTALS								
Sum of Individual VOC	685	470	2998	440	315	466	674	762
Total Organic Carbon	5200	1900	11000	2300	2500	2400	2500	4300

Table 5. Flume Hood Dilution Factors

Date	Injection Room/Hood	Sample Room	Exhaust Duct Concentration	Average Concentration	Dilution Factor
2/28	652B	652	$20.6 \cdot 10^{-9}$	$21.5 \cdot 10^{-12}$	958
3/4	652B	652	$41.3 \cdot 10^{-9}$	$22 \cdot 10^{-12}$	1870
2/28	650B	650	$36.9 \cdot 10^{-9}$	$51 \cdot 10^{-12}$	724
3/4	650B	650	$37.1 \cdot 10^{-9}$	$40 \cdot 10^{-12}$	928
2/28	646A	646	$36.9 \cdot 10^{-9}$	$11 \cdot 10^{-12}$	3354
2/28	646A	644		$32 \cdot 10^{-12}$	1153
3/4	646A	646	$37.1 \cdot 10^{-9}$	$5 \cdot 10^{-12}$	7420
3/4	646A	644		$15 \cdot 10^{-12}$	2473
3/2	626A	626	$33.1 \cdot 10^{-9}$	$13 \cdot 10^{-12}$	2526
3/2	624A	624	$38.2 \cdot 10^{-9}$	$7 \cdot 10^{-12}$	5457
3/2	624A	622		$12 \cdot 10^{-12}$	3183
3/4	624B	624	$38.3 \cdot 10^{-9}$	$5 \cdot 10^{-12}$	7640
3/2	624B	626	$(32.6 \cdot 10^{-9})^{624B}$	$5 \cdot 10^{-12}$	6520
3/3	622A	622	$34.8 \cdot 10^{-9}$	$8 \cdot 10^{-12}$	4350
3/3	620B	624		$7 \cdot 10^{-12}$	4971
3/3	620B	620	$37.1 \cdot 10^{-9}$	$\leq 5 \cdot 10^{-12}$	7420?
3/3	620B	622		$27 \cdot 10^{-12}$	1374
3/4	620B	620	$37.1 \cdot 10^{-9}$	$5 \cdot 10^{-12}$	7420
3/4	620B	622		$15 \cdot 10^{-12}$	2473

Table 6. Results of Flue Gas Re-entrainment Test
Continuous Injection

Injection Rate 2.03 lpm of 1% SF_6
into Combustion Flue of Room 117
Flue Concentration of SF_6 = 57.0 ppm
Flue Flow Rate = 12.5 cfm

Room Air Intake Tracer Concentrations and Dilution Ratios

Intake Room #	Concentration (ppb)	Dilution Ratio
A	136.3	0.002
B	147.3	0.003
C	97.3	0.002
D	0.68	0.000
E	12.4	0.000
Music	2.1	0.000
Gym	104.8	0.002
Corridor	597.0	0.010

Resulting Tracer Concentrations in Rooms after 35 minutes

Room Number	Concentration (ppb)	Quantity of Tracer (cc)	Quantity Dilution Ratio
A	17.4	4.15	0.008
B	21.8	5.2	0.009
C	31.4	8.9	0.016
D	0.7	0.2	0.000
E	1.4	0.4	0.001
Music	1.8	1.0	0.002
Gym	3.4	1.9	0.003

Table 7. Estimate of the Pollutant Levels Caused by Flue Exhaust Re-entrainment

Typical Pollutant Generation Rates of Gas Convective Heaters
(50000 BTU per Hour)

Carbon Dioxide	2.690	kg/hr
Carbon Monoxide	4.374	g/hr
Nitrogen Dioxide	0.632	g/hr
Nitric Oxide	0.896	g/hr

Model Parameters Used to Estimate Re-entrainment

Room	Vol m^3	Dilution Ratio	ACH
K	162.67	0.044	0.63
I	162.67	0.002	0.58
gym	3095.26	0.055	0.41

Predicted Rise in Carbon Dioxide, Carbon Monoxide, Nitrogen Dioxide and Nitric Oxide Caused by Continuous Operation of Heating System 125.

Room	CO_2 mg/m^3	ppm	CO mg/m^3	ppm
K	1154	641	1.9	1.6
I	57	32	0.1	0.1
gym	116	65	0.2	0.2

Predicted Rise in Nitrogen Dioxide and Nitric Oxide Caused by Continuous Operation of Heating System 125.

Room	NO_2 mg/m^3	ppm	NO mg/m^3	ppm	NOx mg/m^3	ppm
K	0.27	0.15	0.38	0.31	0.66	0.36
I	0.01	0.01	0.02	0.02	0.03	0.02
gym	0.03	0.02	0.04	0.03	0.07	0.04

Table 8. HVAC Equipment Room PV Duct Inleakage

Supply = 10854 cfm
Return = 9074 cfm

A Train

Time	Room Concentration (ppb)	Supply Concentration (ppb)	Return Concentration (ppb)	Flow (cfm)
5	30.0	500	92	153
6	28.3	460	92	147
7	26.7	460	118	147
8	25.2	480	120	164

Table 9. Secondary-Containment Test for Chemical Vapor Deposition Process (Orcutt, 1988)

Time (sec)	Sample Location	Measured SF6 Concentration (ppb)	Equivalent Workroom Contamination (ppm)
30	North	0.335	0.7
360	North	10.33	2.8
30	South	0.480	0.11
360	South	9.90	2.7
30	East	5.76	1.5
360	East	50.0	13.4

Table 10. Dilution Ratios in Disassembly Room
Concentration in Supply Conveyor Room = $1.33 \cdot 10^{-8}$

Station	Dilution Ratio
B	0.12
D	0.47
E	0.32
G	0.14
H	0.34
I	0.12

Step 1   Injection

Step 2   Mixing
1/2 - 1 hr

Step 3   Collect Samples

t= 0 min   t=15 min   t=30 min ••• t= ~120 min

130 ppb
Tracer Monitor

100 ppb
Tracer Monitor

78 ppb
Tracer Monitor

18 ppb
Tracer Monitor

On-Site

Step 4

Off-Site

130 ppb
Tracer Monitor

Analyze
Samples

Step 5

Calculate $I = \dfrac{1}{t} \ln \dfrac{C_0}{C_t}$

Tracer Decay

I ~ 1.0 ACH

ln(ppb)

5
4
3

0   30   60   90   120
Time

Equipment

SF_6

Sample
Bags/Syringes

Sample
Pump

Monitor

130 ppb
Tracer Monitor

Figure 1. Schematic of Steps Required to Perform Ventilation Measurements Using the
Tracer Decay Method

General Tracer Measurement System
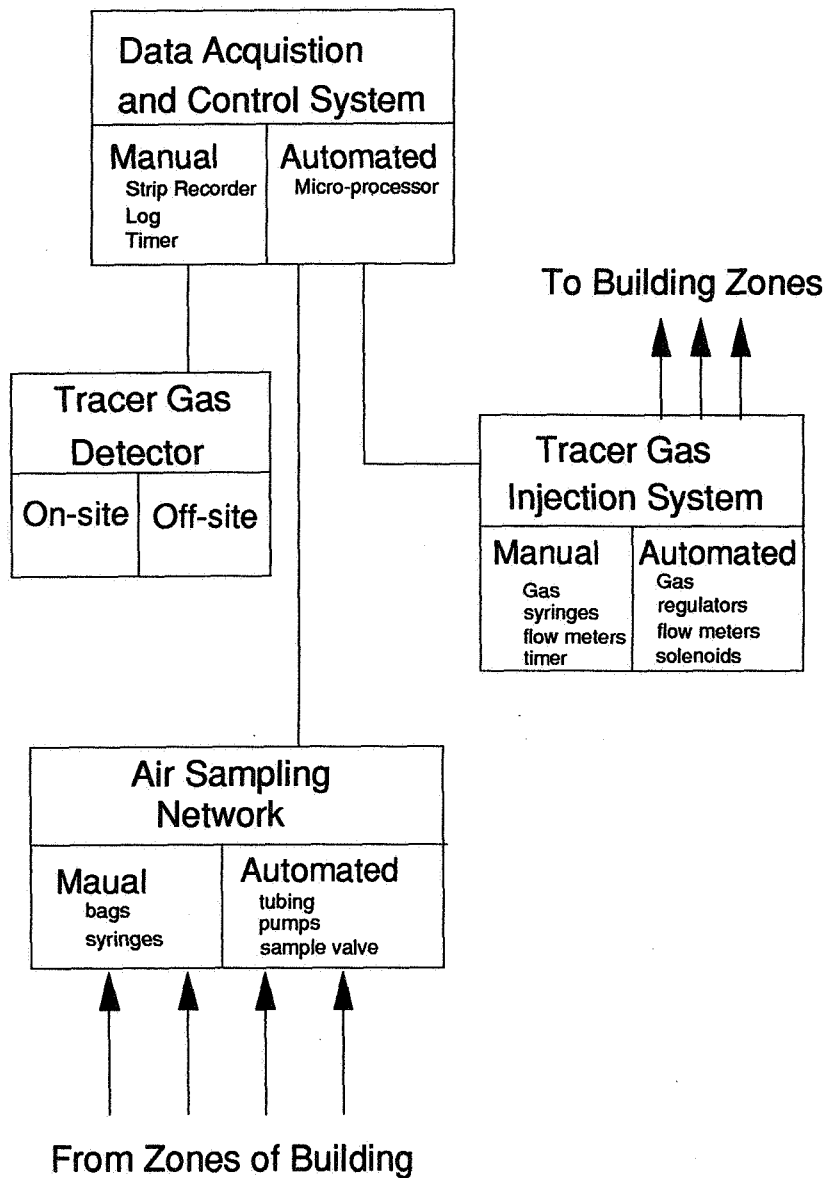
Figure 2. A Schematic of a General Tracer Gas System for Building Ventilation Studies

Schematic of Elementary School

100	101	
102	103	
104	105	
106	Cafeteria	
Entrance Hall	Music	
Office	Gym	

	Volume (cu. ft.)	Tracer Quantity
Classrooms	7500	2 cc
Cafeteria	30000	8 cc
Gym	45000	12 cc
Office	15000	4 cc
Music	7500	2 cc
Hall	3750	1 cc

Target Tracer Concentration
10 ppb SF_6
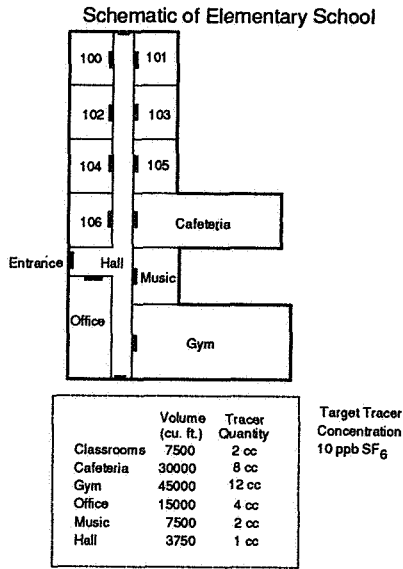
Figure 3. An Example of a Compartmentalized Building - An Elementary School

Ventilation - Room 101
AI = 0.33 ACH (0.03)

Ventilation - Room 105
AI = 0.66 ACH (0.21)

Ventilation - Cafeteria
AI = 0.92 ACH (0.06)

Ventilation - Gym
AI = 0.38 ACH (0.01)
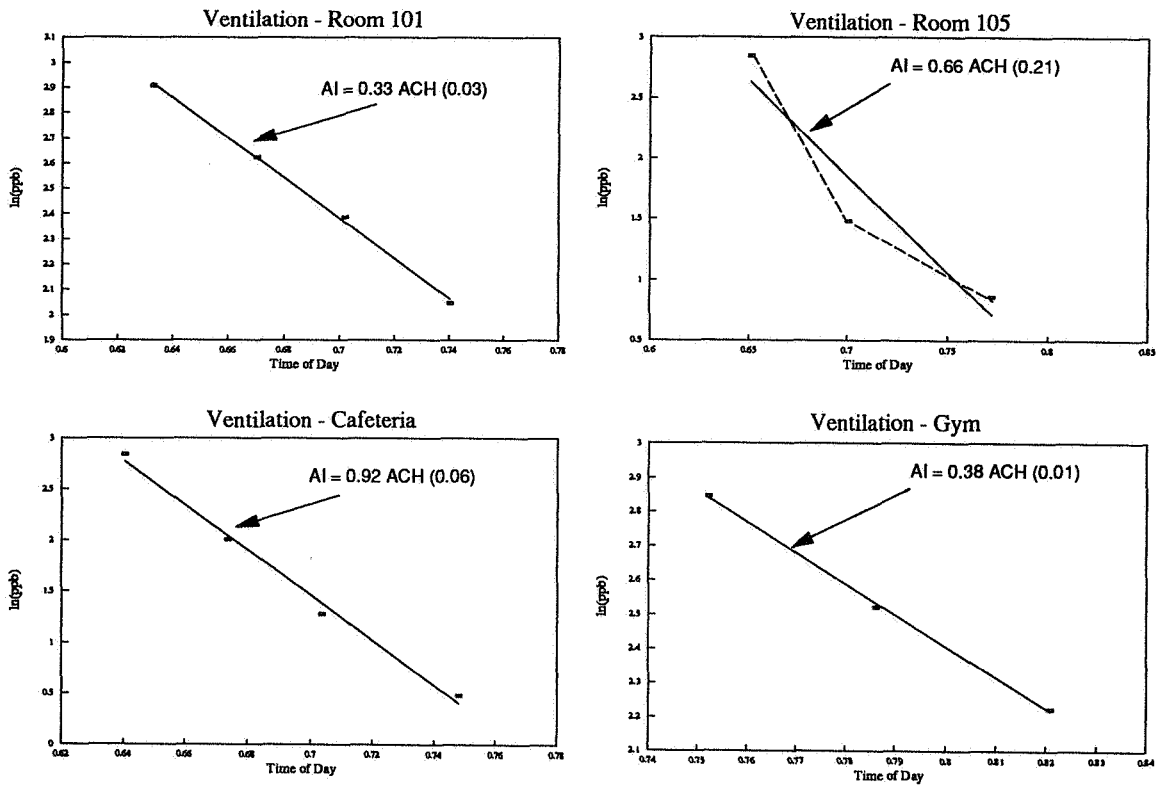
Figure 4. Results of a Tracer Decay Test in Four Rooms of an Elementary School
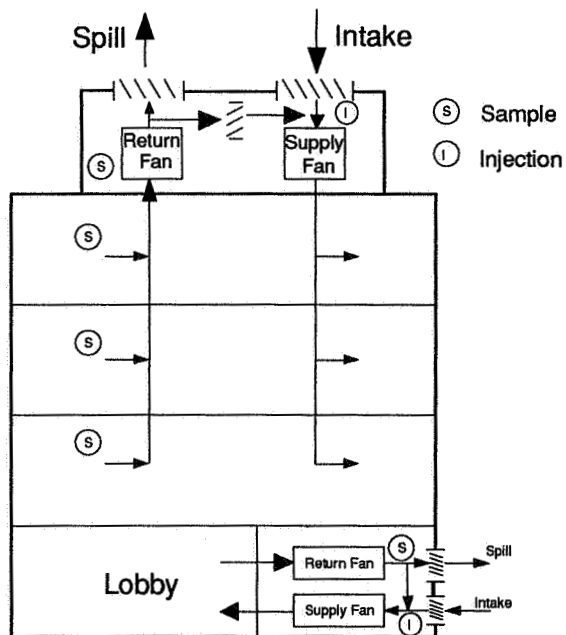
Schematic of Building Ventilation System

Figure 5. A Schematic of a HVAC System Used for Tracer Sampling and Injection in a Large Building
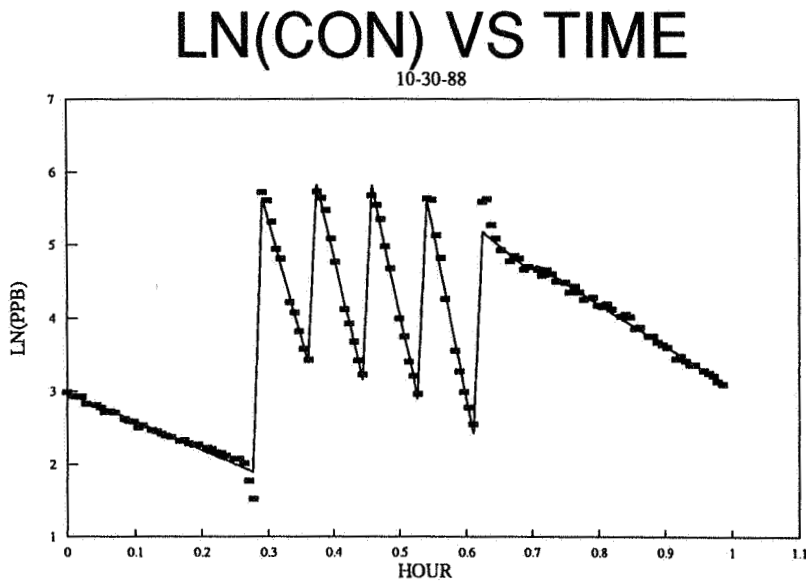
Figure 6. Results of a Tracer Decay Test Using an Automated Tracer Monitoring System
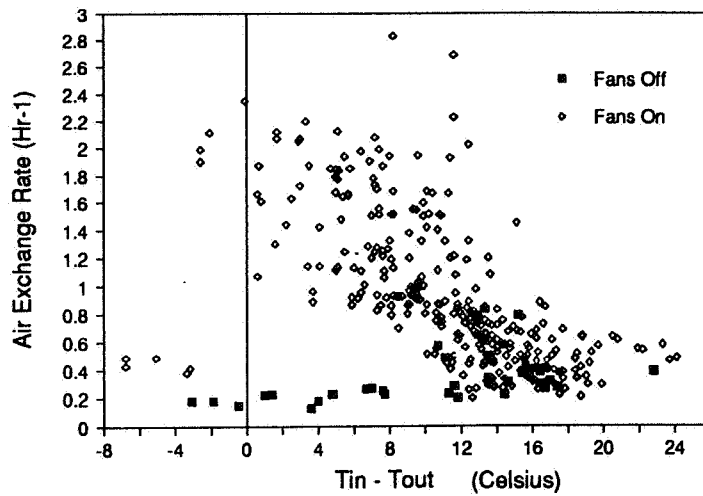
Figure 7. Air Exchange Rate versus Inside-Outside Temperature Difference
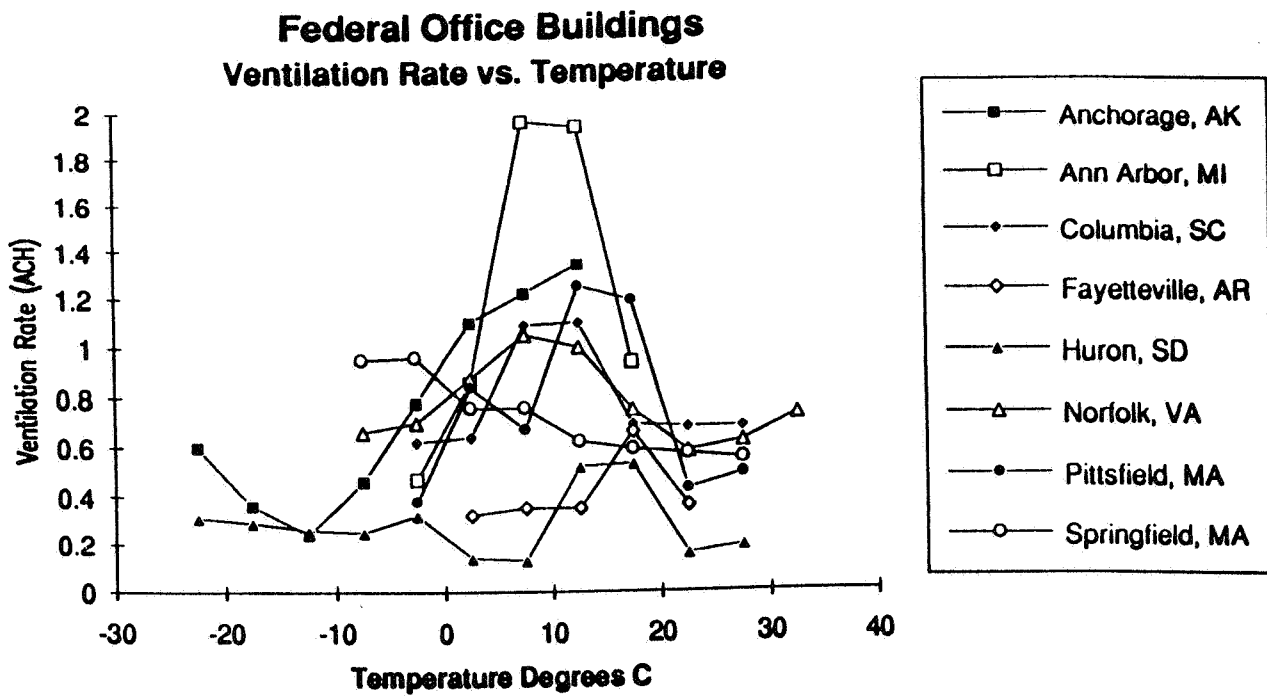
Figure 8. Example of Weather Dependence of Ventilation Rates in Eight Federal Office
Buildings

Air Handling System
with recirculation
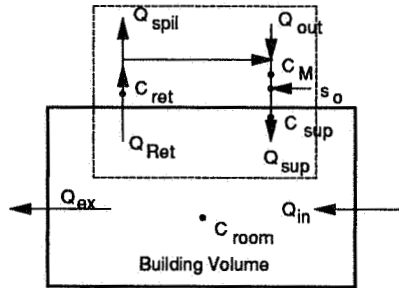
Equations

Percent Outside Air

$$\% = 1 - \frac{C_M}{C_{Ret}}$$

Air Infiltration

$$Q_{in} = Q_{sup}\left(\frac{C_{sup}}{C_{room}} - 1 \right)$$

Supply Airflow

$$Q_{sup} = \frac{s_0}{C_{sup} - C_M}$$

Figure 9. Schematic of Measurement Locations for Evaluation of HVAC System

Duct Flow Measurement

Measure
Up Stream

Sample Tube

Inject Trcer
Down Stream

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2 3

Tracer Monitor

Tracer
Flow
Controller
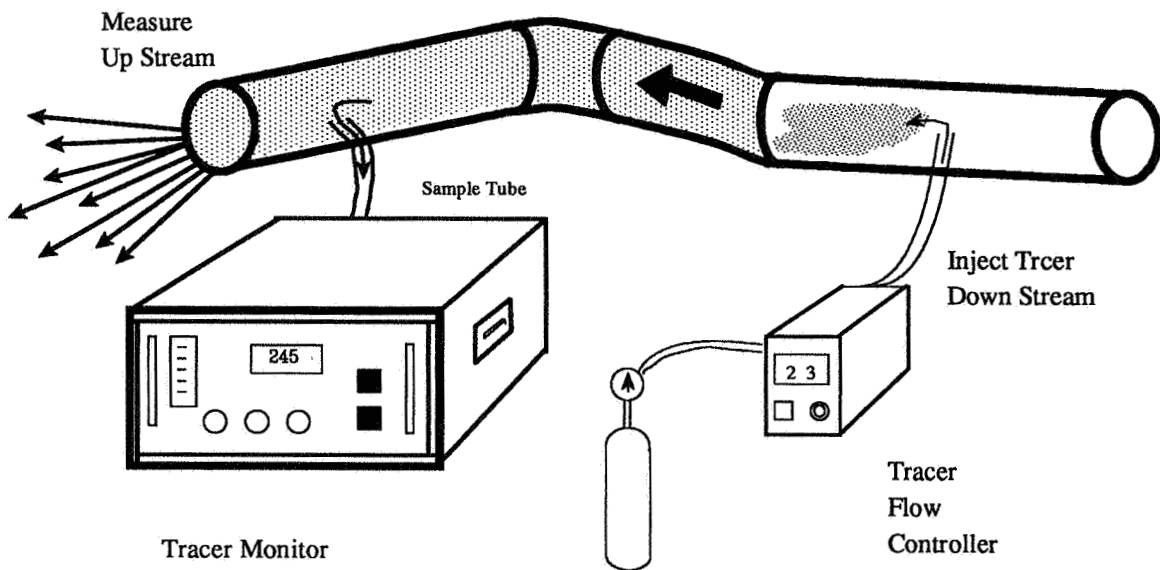
Figure 10. Use of the Constant Injection Method for Measuring Airflow in a Ventilation Duct
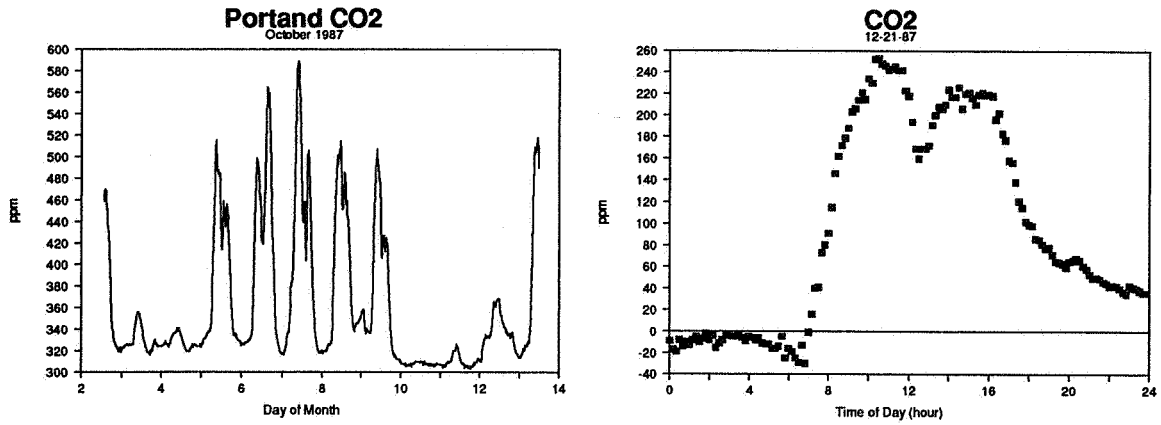
Figure 11. Daily Variation in Carbon Dioxide Levels in an Office Building
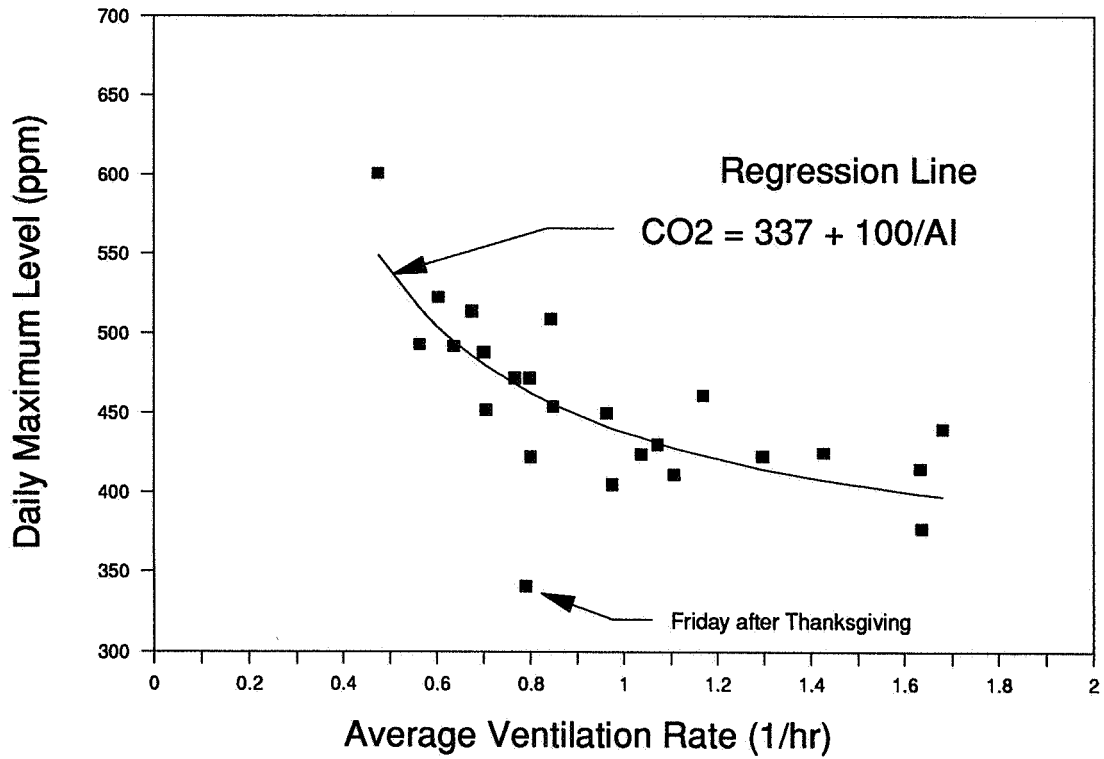
Figure 12. Carbon Dioxide Level versus Ventilation

Total Organics versus Air Exhange

Portland East Federal Building
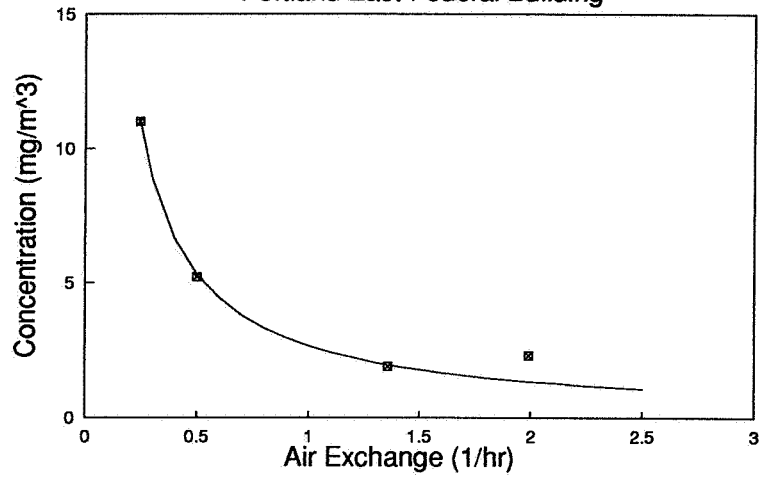
Figure 13. Total Volatile Organics versus Ventilation Rate
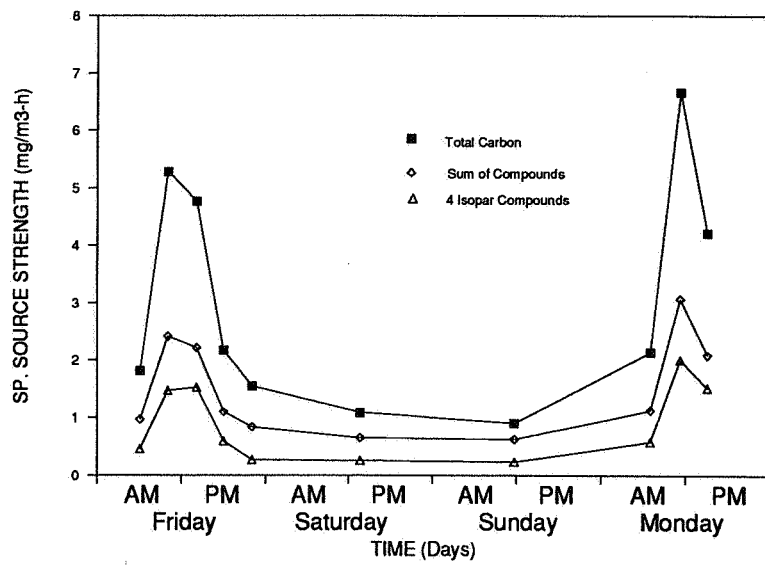
Figure 14. Variation in VOC Sources over a Four Day Period

MULTIPLE TRACER GAS CHROMATOGRAPH SEPARATION
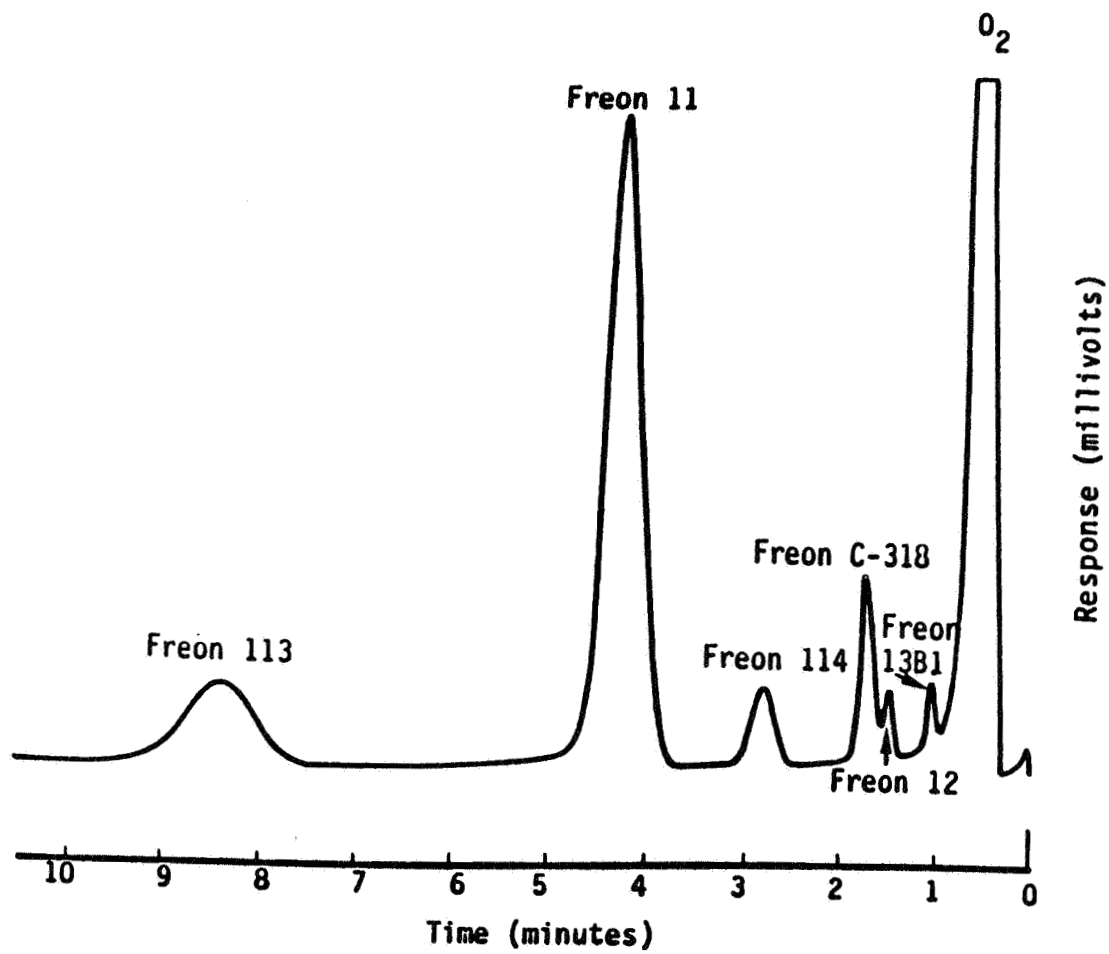
Figure 15. Separation of Multiple Tracers on a Gas Chromatograph
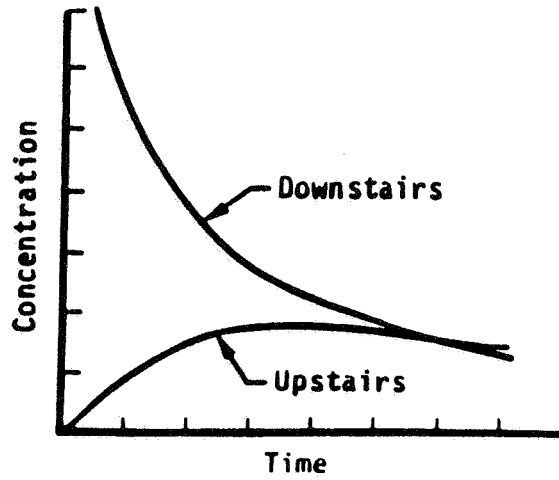
Concentration of Freon 114 against time, upstairs and downstairs.
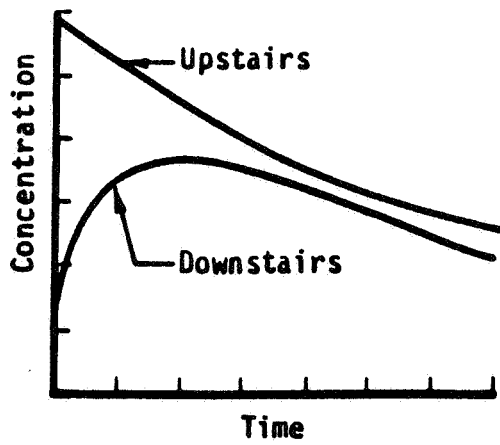
Concentration of Freon 12 against time, upstairs and downstairs.

Figure 16. Example of Two Tracer Decay Test
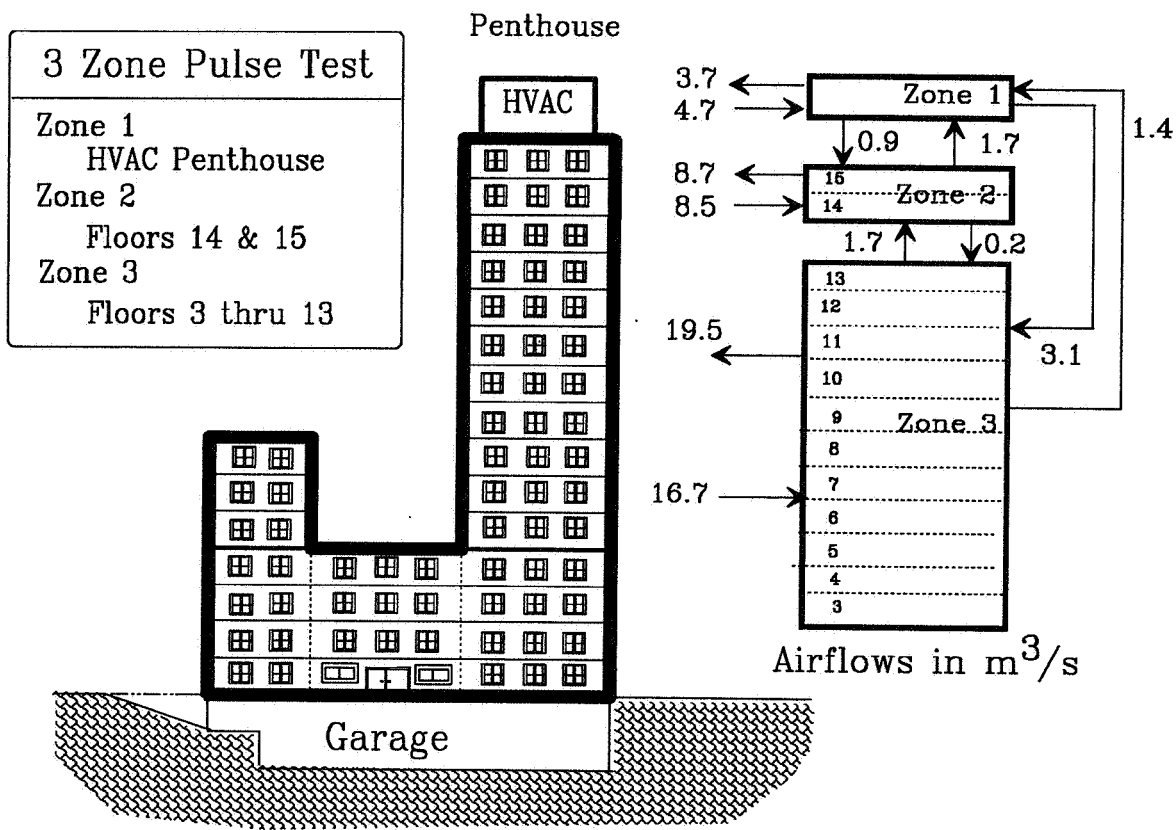
Figure 18. Use of Single Tracer to Determine Air Change of   Sections of a Building

Multi-Tracer Sandwich Test
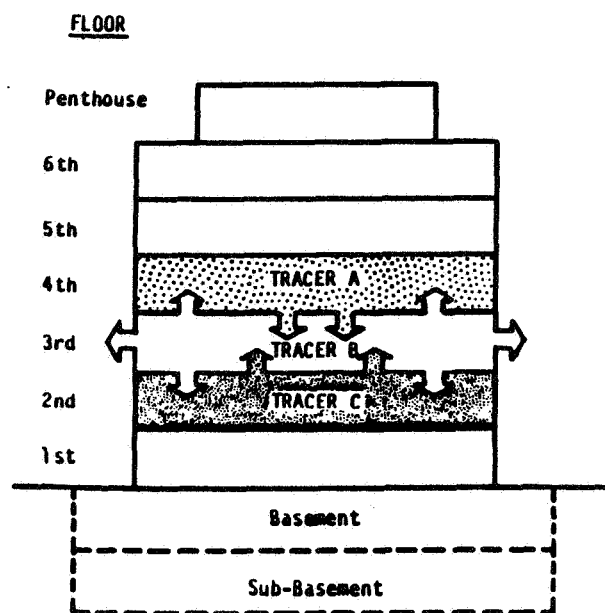
FLOOR

Figure 17. Use of Multiple Tracers to Measure the Ventilation Rate on a Floor of a Building
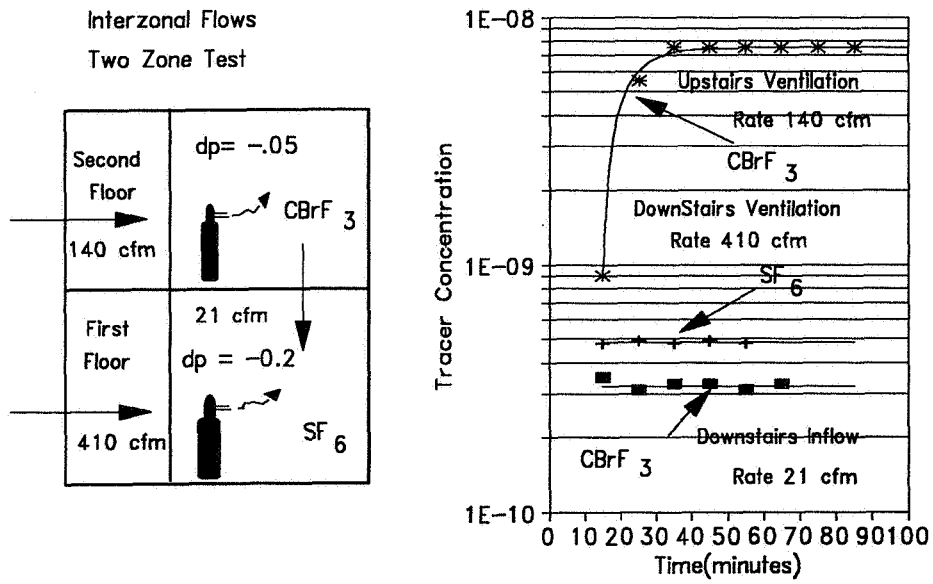
Figure 19. Two Tracer Test for Measuring Airflows from a Laboratory
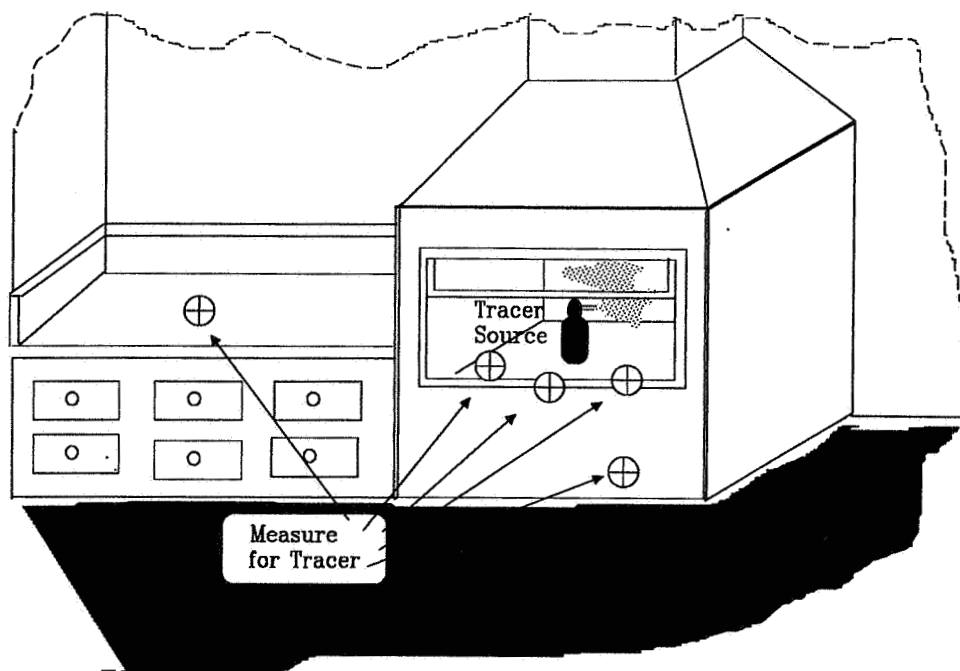
Figure 20. Schematic of Test Procedure for Evaluating Fume Hoods

Re-entrainment
Flow
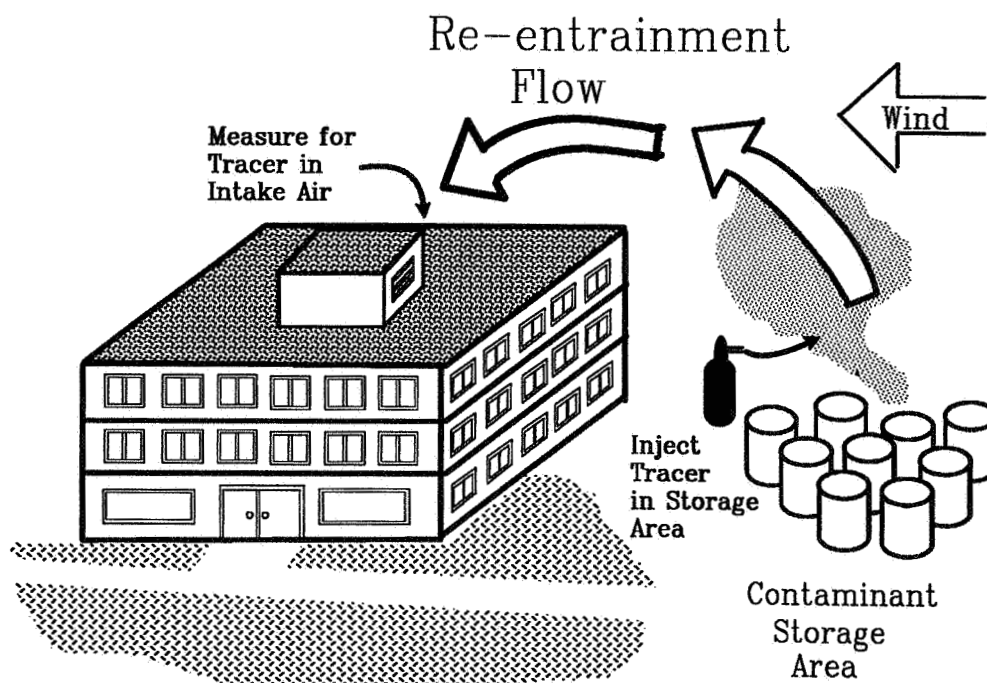
Figure 21. Schematic of Test for Measuring Re-entrainment and Re-circulation from an
Exterior Contaminant Source
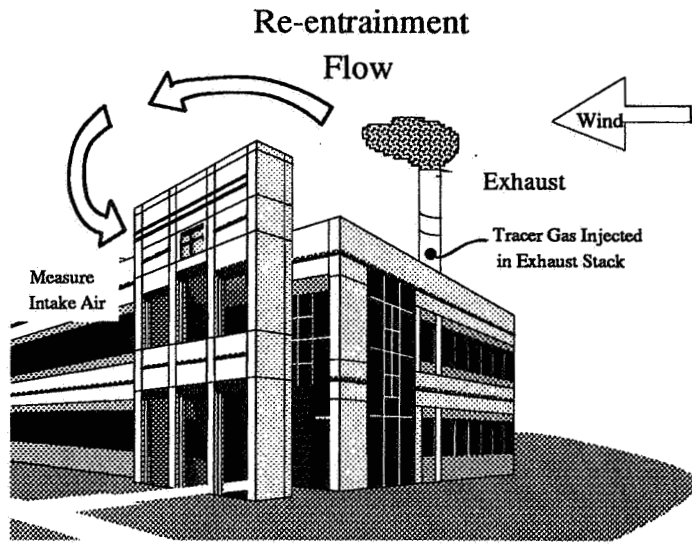
Figure 22. Schematic of Test for Measuring Re-entrainment and Re-circulation from a Building Exhaust
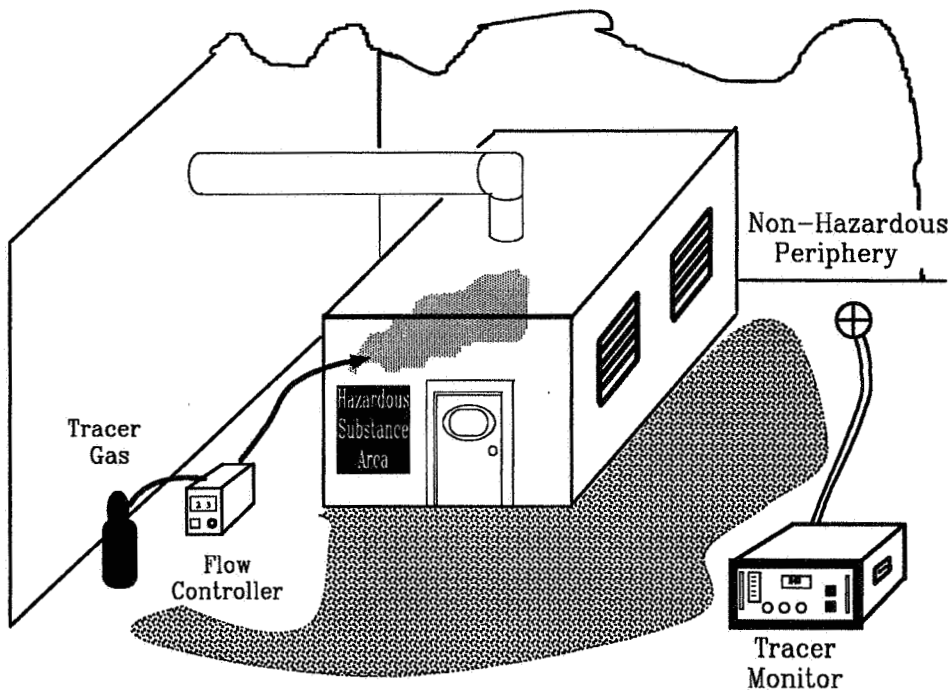
Figure 23. Integrity Breach Testing Using a Tracer Gas

F is Ventilation Rate of Control Room

C_e is equilibrium tracer in the control room

C_p is tracer concentration in periphery

$$\text{Inflow} = F \frac{C_e}{C_p}$$
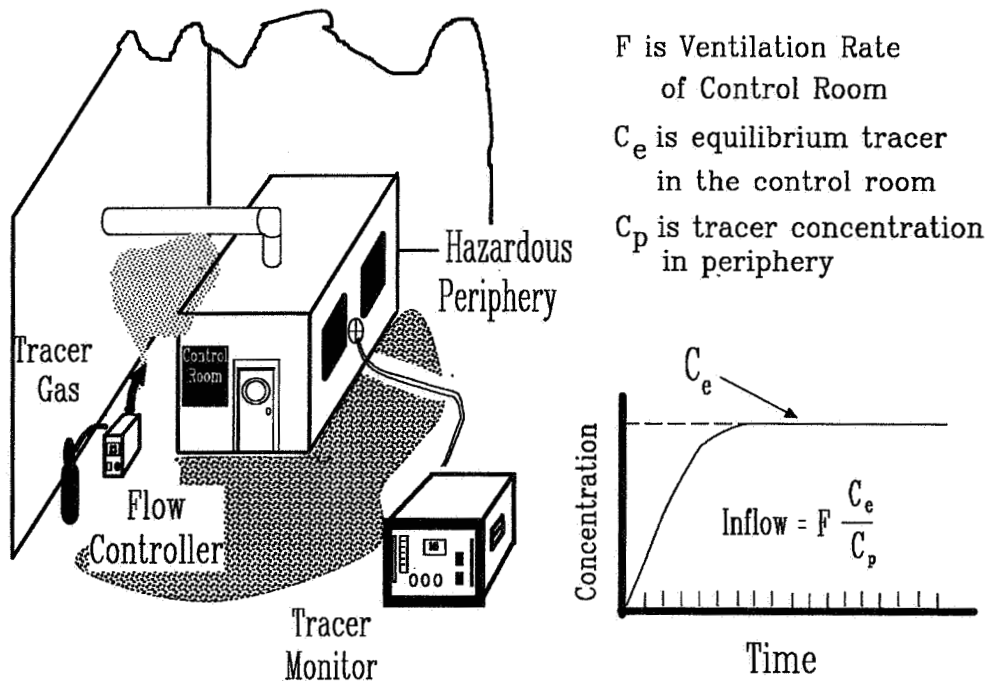
Figure 24. Testing for the Potential of Contaminant Leakage into a Control Room.

$\triangle C \approx \text{Inflow Leakage}$
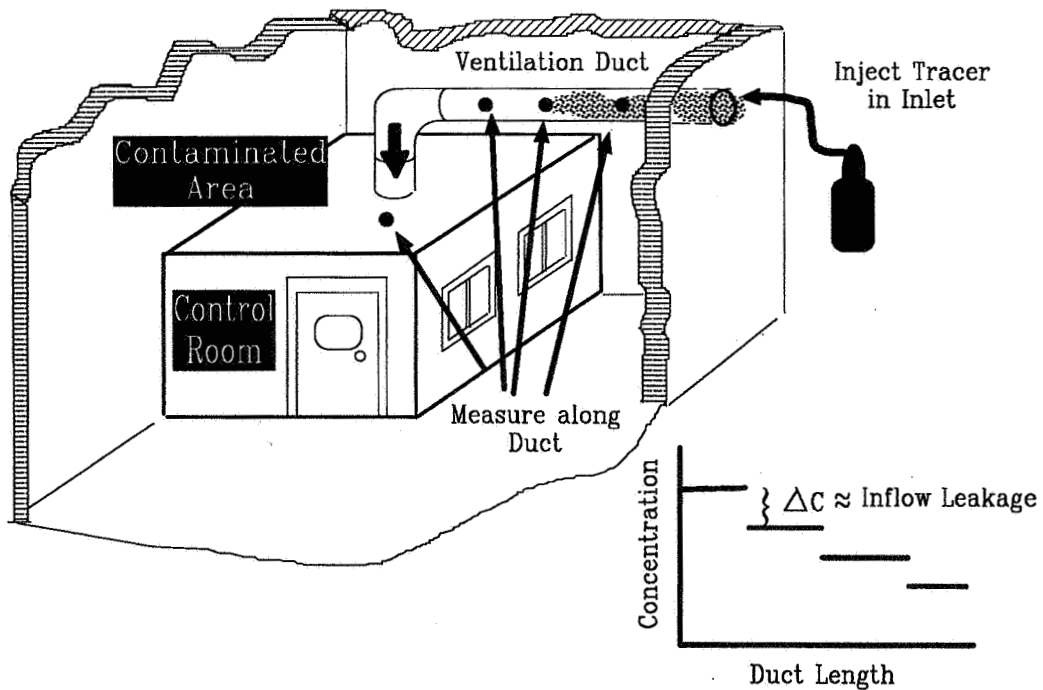
Figure 25. Measuring and Locating Leaks in Negative Pressure Supply Air Duct Passing through a Containment Area.

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Cross Section View

Top View of Roof
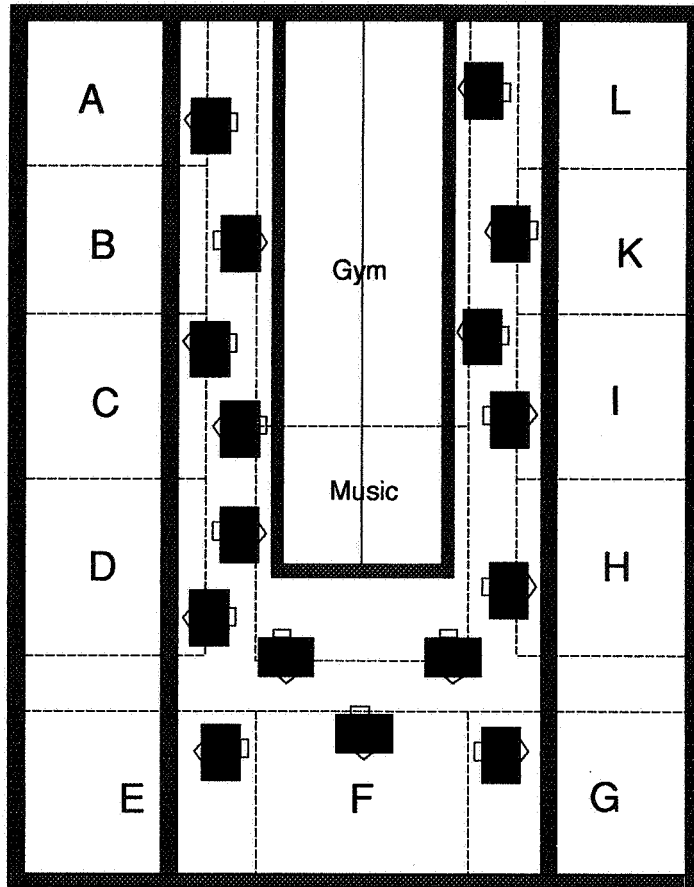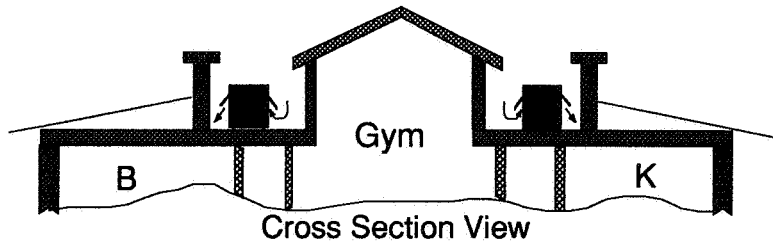
Flue Exhaust Re-entrainment

Figure 26. Schematic of Flue and Intake Locations for Tested Elementary School.

Figure 27. Schematic of Flows Used to Quantify Inleakage Data
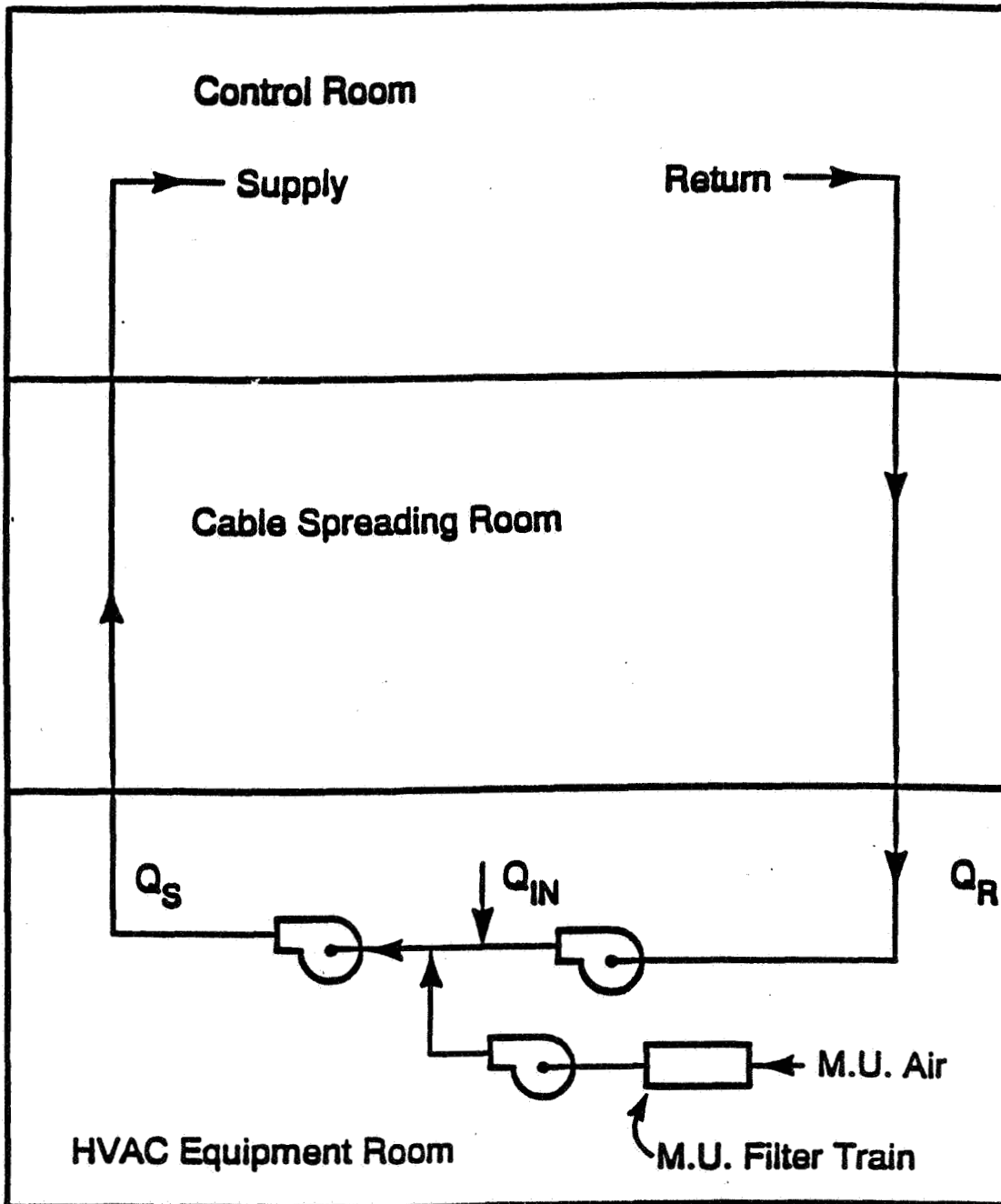
Figure 28. Plan View of Disassembly Room
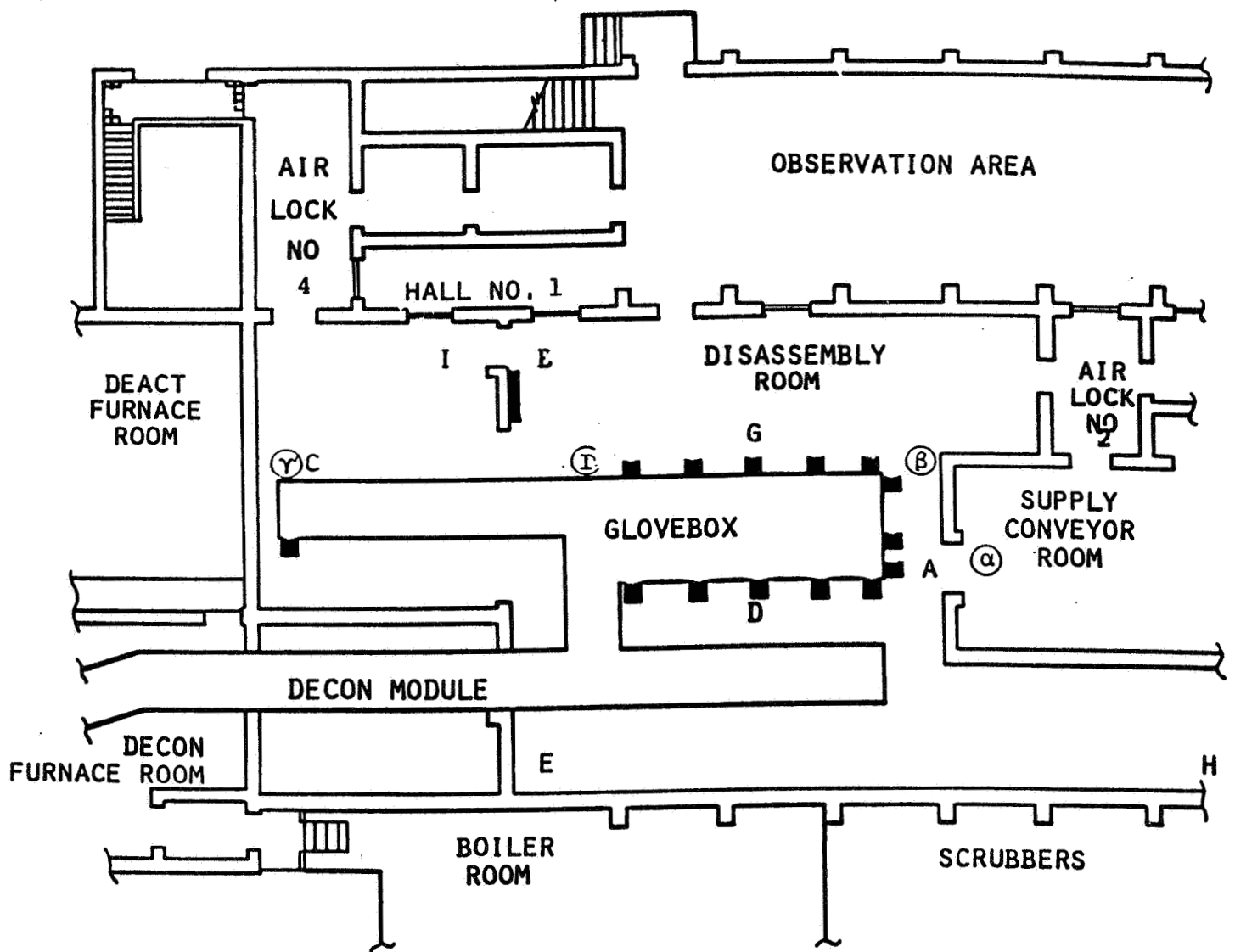
Figure 29. Schematic of Test Tracer Test in a Nuclear Facility.

Solid lines are design airflow paths, dashed lines are measured airflow paths.
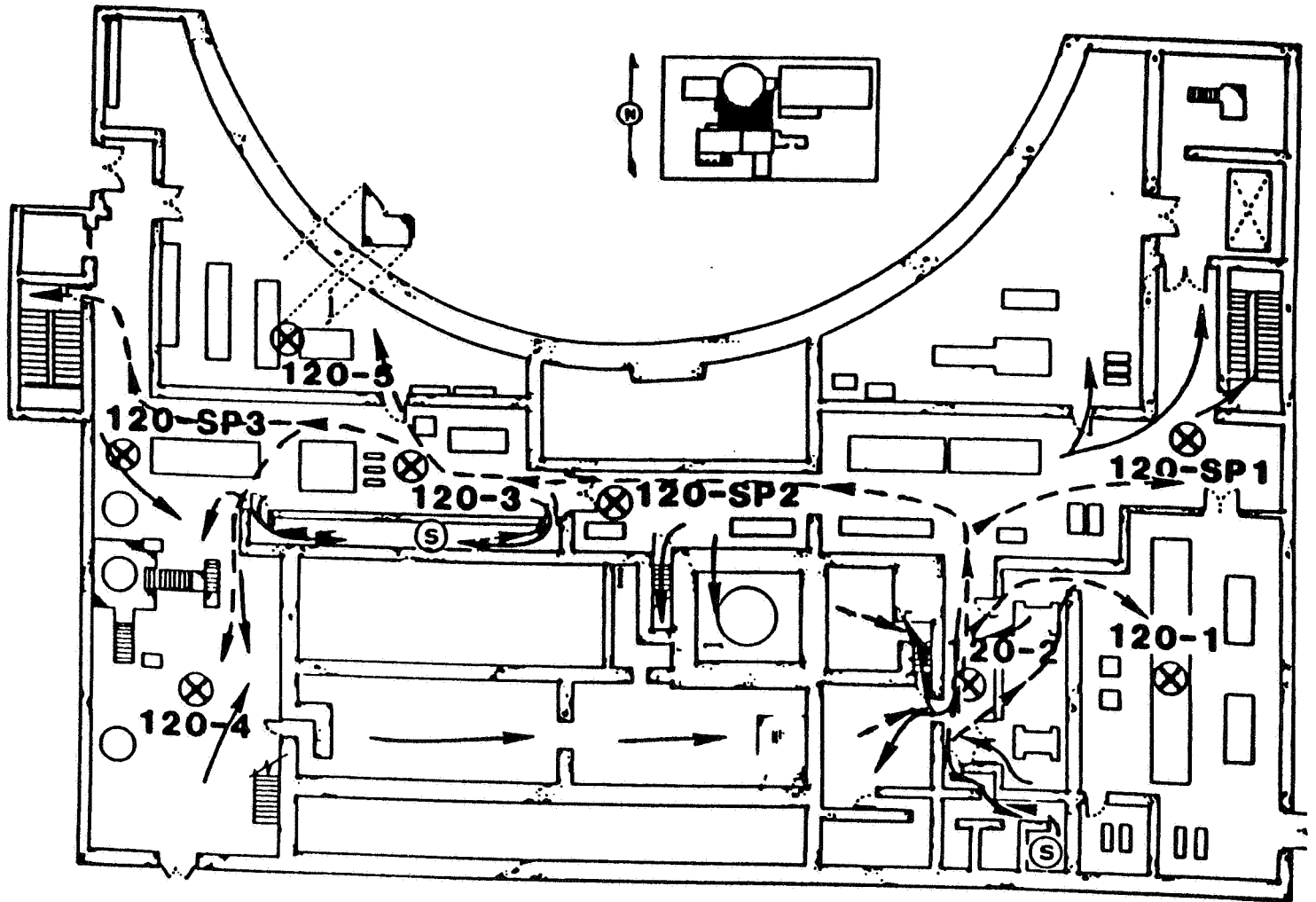
Figure 30. Expanded View of Design and Measured Airflow Paths in a Nuclear Facility.
Solid lines are design airflows paths, dashed lines are measured airflow paths.
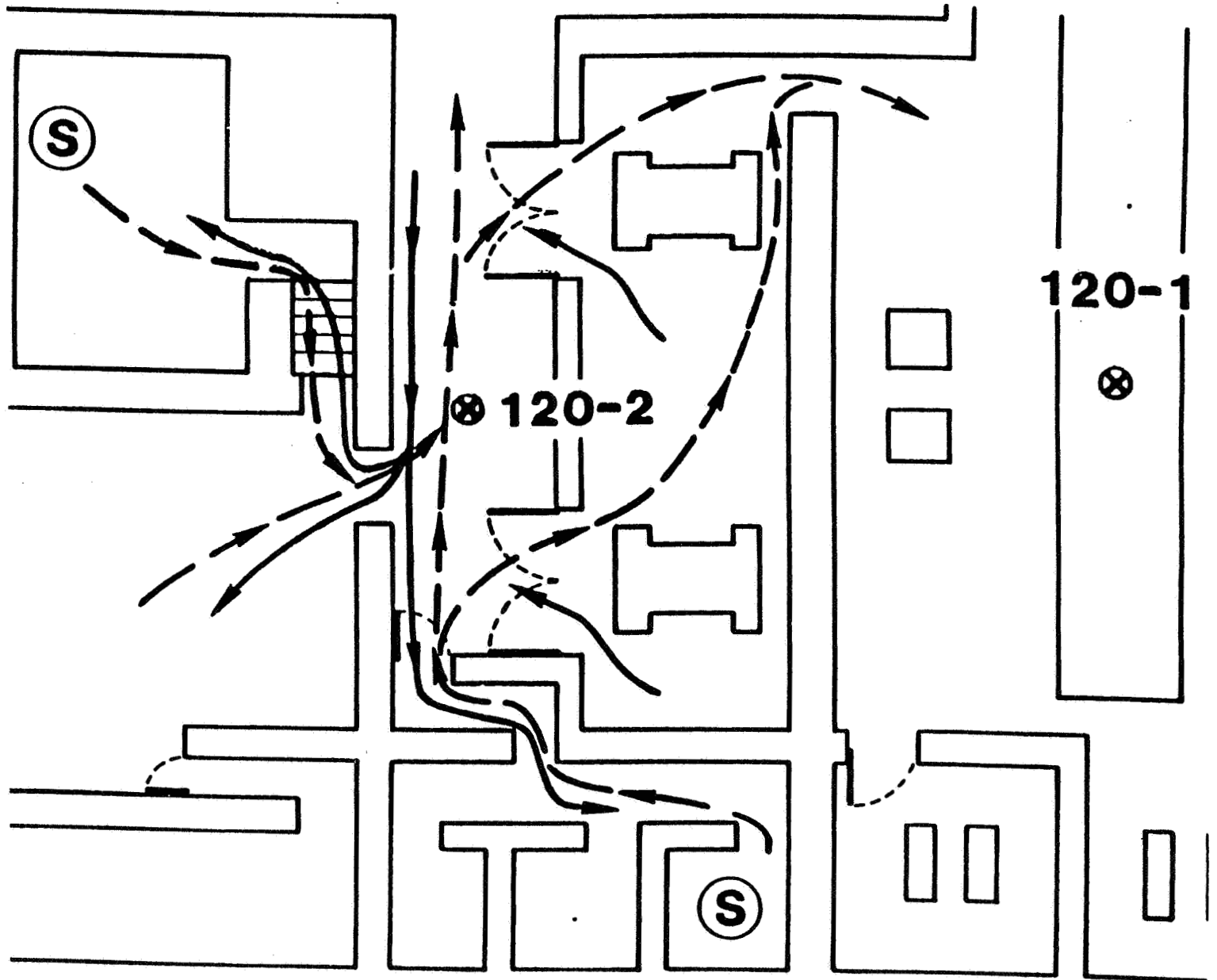
Figure 31. Example of Steady State Tracer Concentrations From Which Dilution Ratios Can be Calculated
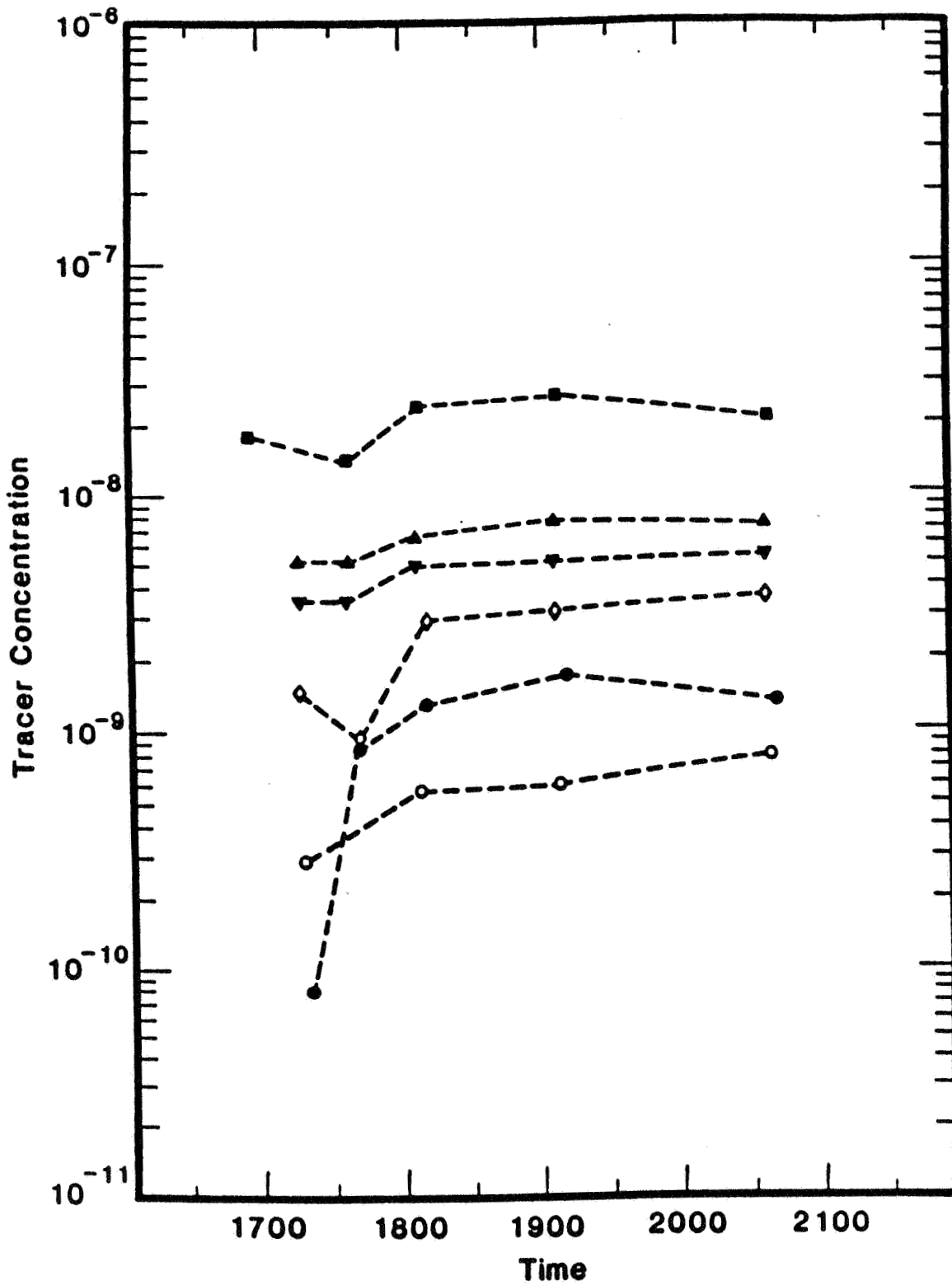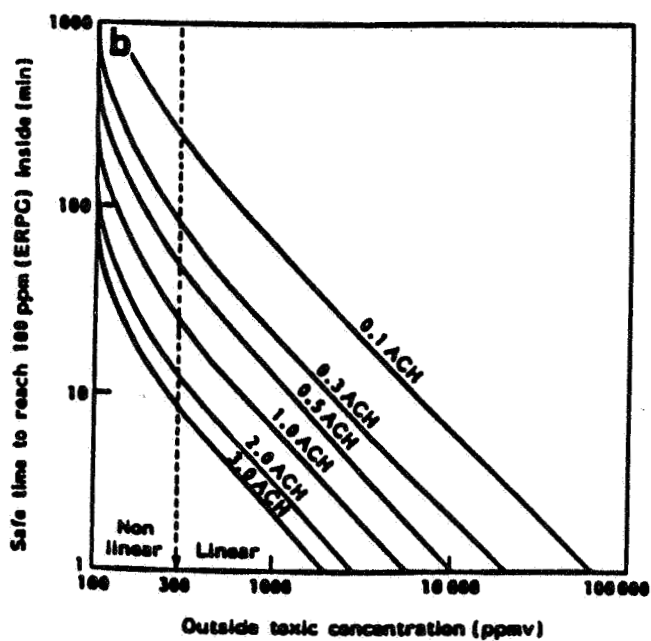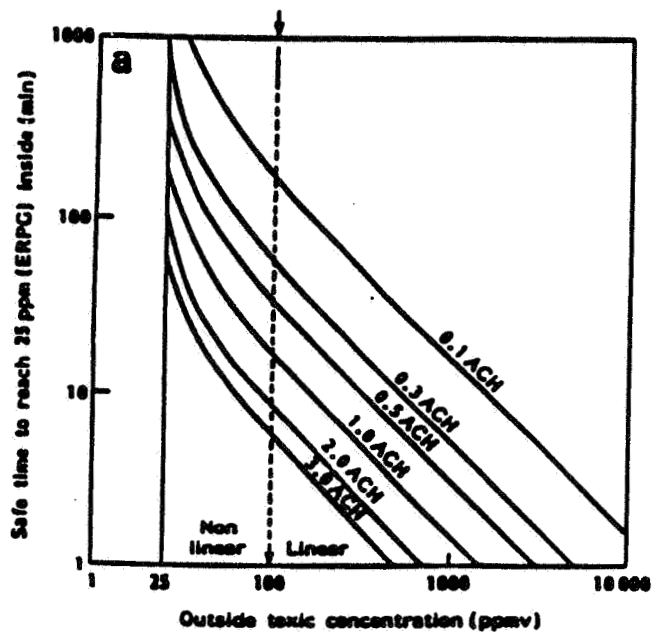
Figure 32. Nomographs Used to Determine Occupancy Time Versus Toxic Challenge for
Temporary Safe Havens (Jann, 1989)

Appendix I
Theory of Tracer Gas Methods for Determining the Building Air Change Rate and the Building Airflow Rate

The theoretical assumptions which form the basis for tracer gas testing of single zone buildings can be explained by studying Figure 1. In this situation, there is one well mixed zone in which air is entering through known or unknown flow paths at a flow rate Q_{in} and exiting the space through know or unknown flow paths at a flow rate Q_{out}. If a tracer gas is introduced into the space, the basic equation for a tracer gas concentration C(t) in the zone is given by the conservation of mass of the tracer in the zone:

$$\frac{d}{dt}(\rho VC) = C_0 Q_{in} - C Q_{out} + s \tag{1}$$

where

ρ is the density of air in the zone (moles of air per m^3)

V is the volume of the zone

C is the concentration of the tracer per unit mass of air (moles of tracer gas per mole of air)

C_0 is the concentration of tracer in the region exterior to the zone in the same units as C

Q_{in} is the mass flow rate of air into the zone from the exterior (moles of air per second)

Q_{out} is the mass flow rate of air out of the zone (moles of air per second)

s is the generation rate of tracer in the zone (moles of tracer per second)

V is the volume of the zone in m^3.

The basic assumptions leading to the derivation of equation (1) are:

- The tracer gas in the zone is well mixed and therefore can be represented by a concentration density independent of the spatial position in the room.

Note: This assumption can be relaxed somewhat to the assumption that the average concentration in the zone is the same as the average value of the tracer in the airflows out of the zone

- The air in the room is of uniform density which is independent of the spatial position in the room.

Note: To this point, no assumption has been make as to the temporal dependence of the quantities in equation (1). That is ρ, C, C_0, Q_{in}, Q_{out}, s and even V are arbitrary functions of time as long as the above two assumptions are satisfied. Useful and accurate information can be obtained from tracer gas experiments without restrictive assumptions on the temporal dependence of this quantities.

The second basic equation for analyzing tracer gas data is the equation for conservation of mass for air flowing into and out of the zone. Under the second assumption of equation (1), the conservation of mass for air flowing into and out of a zone is:

$$\frac{d}{dt}(\rho V) = Q_{in} - Q_{out} \tag{2}$$

In the common situation of constant temperature, pressure and zone volume, equation 2 is nothing more than a statement that the flows into the zone are equal to the flows out of the zone. Equations (1) and (2) have been written in mass units: flow in mass per unit time and concentrations in mass of tracer per mass of air. This is the simplest form of the equations when temperature and pressure are not constant or when the exterior temperature is different from the interior temperature. It is also in the authors' opinion, the easiest way to make temperature and pressure corrections when such precision is required; however in most investigations little error is introduced by using volume flows rates (cfm) or volume concentrations. If the left hand side of equation (1) is differentiated by parts and equation (2) substitutes for $\frac{d}{dt}(\rho V)$, equation (1) reduces to

$$\rho V \frac{dC}{dt} = (C_0 - C)Q_{in} + s \tag{3}$$

Equation (3) is the fundamental equation of tracer gas test methods in a single well mixed zone..

There are several quantities of interest which can be determined by tracer gas experiments:

- The air change rate $\lambda(t)$ defined as

$$\lambda = \frac{Q_{in}}{\rho V}$$

- The air inflow rate $Q_{in}(t)$

Equation (3) can be put into several different forms depending on which quantity is to be measured and the type of tracer experiment (decay test, constant injection or constant concentration) to be performed.

Determination of the Air Change Rate λ

Equation (3) can be solved for λ to yield

$$\lambda(t) = -\frac{d}{dt}(\ln \hat{C}) + \frac{\hat{s}}{\rho V \hat{C}} \tag{4}$$

where

$$\hat{s} = s - \rho V \frac{dC_0}{dt}$$

$$\hat{C} = C - C_0$$

Though in principle, equation (4) can be used to estimate $\lambda(t)$, in practice accurate determination of the derivative of the concentration from experimental data is not possible. In practice one is also usually interested in the average air change rate over a period of time (t_1, t_2). Integrating equation (4) over the interval (t_1, t_2) leads to the equation:

$$\lambda_{av}(t_1, t_2) = \frac{1}{t_2 - t_1} \ln\left(\frac{\hat{C}(t_1)}{\hat{C}(t_2)}\right) + \frac{1}{t_2 - t_1} \int_{t_1}^{t_2} \frac{\hat{s}(\tau) d\tau}{\rho(\tau) V(\tau) \hat{C}(\tau)} \tag{5}$$

where

$$\lambda_{av}(t_1, t_2) \equiv \frac{1}{t_2 - t_1} \int_{t_1}^{t_2} \lambda(\tau) d\tau$$

Equation (5) can be used to determine the building air exchange rate in all three tracer test methods. All quantities on the right hand side of equations (5) are measurable during a tracer test. However, each tracer test method is designed to simplify the form of equation (5) and it is the simplified forms which are usually used in the analysis of tracer data. Equation (5) is useful in assessing errors caused by the lack of fulfillment of the assumptions of each test procedure: the effect of small leaks of tracer in a tracer decay test, the error caused unsteady flows or lack of equilibrium in the constant injection test and the error caused by deviations in the zone concentration from the target concentration in the constant concentration method.

The Tracer Decay Method.
For the tracer decay method, after injection, the tracer injection rate $s = 0$. The exterior tracer concentration must also be constant (for a properly selected tracer, it is zero). In this case the equation (5) reduces to the following simple relationship for the building air change rate which occurred in the time period (t_1, t_2):

$$\lambda_{av}(t_1, t_2) = \frac{1}{t_2 - t_1} \ln\left(\frac{\hat{C}(t_1)}{\hat{C}(t_2)}\right) \tag{6}$$

In practice, tracer decay data are usually analyzed by plotting the logarithm of the concentration versus time (see Figure 6 for an example) and portions of the data for which the decay is linear are analyzed statistically by fitting a straight line to the data. That the resulting plot is a straight line requires the added assumption that the flow rates are constant. This is not required for the use of equation (6); however it is necessary to assure that the lack of a logarithmic decay, if it does occur, is not due to poor initial mixing or the invalidity of the single zone assumption.

Constant Injection Method
In the constant injection method, tracer gas is injected into a test volume at a constant injection rate s_0 and the tracer concentration is monitored until an equilibrium value C_∞ is reached. Equations (5) and (6) then reduce to the following simple expression for the ventilation or airflow rate into the test volume:

$$q_{in} = \frac{s_0}{(C_\infty - C_0)} \tag{7}$$

Equation (7) is also used to determine ventilation flow rates in ducts, though the derivation in this case is different and requires the assumption the tracer concentration is uniform across a duct cross-section. It must be emphasized that equation (7) is valid only for steady flows. If fluctuations in the flow are observed (by noticing oscillations in the tracer concentration), equation (5) can be used to obtain the average flow rate during a period of time or to estimate the error caused by using equation (7).

Multi-Chamber Theory

In some investigations, a single zone model of a building is not adequate and a multi-chamber or multi-zone approach must be used. Figure 2 depicts the air and tracer gas fluxes for three interacting zones. The theory behind multi-chamber methods is complex and it is only outlined here. The mass balance for a tracer in the ith zone is given by the equations:

$$\frac{d\rho_i V_i C_i}{dt} = -Q_i^{(out)} C_i + \sum_{i \neq j} Q_{ij} C_j + s_i \quad i = 1...N \tag{8}$$

where $Q^{(out)}_i$ is the sum of all flows out of the ith zone and Q_{ij} is the flow to the ith zone for the jth zone. In the above equations, the exterior denoted by the subscript 0 and N is the number of zones.

Using the conservation of mass for the airflows, equations (8) can be written as

$$\rho_i V_i \frac{dC_i}{dt} = -Q_{i0}(C_i - C_0) + \sum_{\substack{i \neq j \\ j \neq 0}} Q_{ij}(C_j - C_i) + s_i \quad i = 1..N \tag{9}$$

Equation (9) is the basis for most multi-zone and multi-tracer methods. For multi-tracer methods, an equation of the form of (8) or (9) is formed for each tracer gas used in each zone. In principle, in a building with N zones, a properly designed experiment with N tracers can determine all interzonal airflows and the airflows to and from the exterior for each zones (that is, there are N^2 equations for N^2 unknowns). The simplest multi-tracer test is done by injecting a different tracer at a constant injection rate into each zone and waiting until equilibrium is reached. Therefore the derivatives on the left hand side of equation (9) are zero. In the case of strongly interacting zones, this interaction sometimes leads to poorly conditioned equations which are difficult to solve accurately.

To avoid ill-conditioned equations, the system of equations (9) is often integrated over a period of time (t_1, t_2), assuming that the airflows are constant, to produce

$$\rho_i V_i (C_i(t_2) - C_i(t_1)) = -Q_{i0} \int_{t_1}^{t_2} (C_i(t) - C_0(t))dt + \sum_{\substack{i \neq j \\ j \neq 0}} Q_{ij} \int_{t_1}^{t_2} (C_j(t) - C_i(t))dt + \int_{t_1}^{t_2} s_i(t)dt \tag{10}$$

Equations (10) are the bases of the so-called integral-pulse method (see Persily and Axley [1990]) in which pulses of tracer are injected into each zone. One advantage of this method is that all quantities appearing in equations (10) can be measured simply without resorting to the mathematical evaluation of the integrals. The integral of the concentrations are determined using standard integral sampling techniques such as filling air sample bags at a constant rate over the time period of the test. The integral of the injection source rate is the total quantity of tracer injected during the time of the test.

Constant Concentration Method
In a multi-chamber test, if a method can be found to keep the concentrations in all interior zones the same, the derivatives on the left hand side of equation (9) are zero as are the terms in the summation on the right hand side. Equations (9) then yield the following simple expression for the flow of exterior air into the zone:

$$Q_{i0}(t) = \frac{s_i(t)}{(C_{target} - C_0)} \tag{11}$$

where $Q_{i0}(t)$ is the airflow rate into the space from the exterior, C_{target} is the controlled level of tracer and $s_i(t)$ is flow rate of tracer required to maintain the target concentration at the time t. Sophisticated micro-processor controlled systems have been designed by various research laboratories to make constant concentration measurements.

Bounds for $\lambda_{av}(t_1, t_2)$

Thought equation (5) can be used to estimated the average air change for an arbitrary tracer experiment, usually simplified expressions are employed. Bounds on the accuracy of these estimates can be derived from equation (16) for arbitrary injection rate s(t) if either the exterior concentration C_0 is constant or a decreasing function of time.

If c_m is an arbitrary constant concentration and similarly for ρ_m, V_m then by it is straight forward to show from equation (5) and the definition of $s_{av}(t_1,t_2)$ that

$$\lambda_{av}(t_1,t_2) - \frac{1}{t_2-t_1}\ln\left(\frac{\hat{C}_1}{\hat{C}_2}\right) - \frac{1}{t_2-t_1}\frac{s_{av}(t_1,t_2)}{\rho_m V_m C_m} = \frac{1}{t_2-t_1}\int_{t_1}^{t_2} s(\tau)\left(\frac{1}{\rho(\tau)V(\tau)C(\tau)} - \frac{1}{\rho_m V_m C_m}\right)d\tau \quad (12)$$

Let V_{max} and c_{max} denote the maximum values of V and c in the interval (t_1,t_2) and V_{min} and c_{min} the corresponding minimum values of these quantities. Then setting V_m and c_m equal to V_{max} and c_{max} in equation (12), if the exterior concentration of tracer is constant or decreasing, the integral on the right hand side is greater than or equal to zero. Similarly setting V_m and c_m equal to V_{min} and c_{min} in equation (12), the integral on the right hand side of equation (12) is less than or equal to zero. Therefore $\lambda_{av}(t_1,t_2)$ can be bounded by:

$$\frac{s_{av}(t_1,t_2)}{\rho_{max}V_{max}C_{max}} \leq \lambda_{av}(t_1,t_2) - \frac{1}{t_2-t_1}\ln\left(\frac{\hat{C}_1}{\hat{C}_2}\right) \leq \frac{s_{av}(t_2,t_1)}{\rho_{min}V_{min}C_{min}} \quad (13)$$

Application to Constant Injection

For constant injection $\hat{s}(t) = s_0$, a constant value if the exterior tracer concentration is constant. The general expression of equation (9) is valid with $s_{av}(t_1,t_2) = s_0$. In a constant injection experiment two situations are common: case 1 in which the increase in tracer concentration is monotonic and case 2 in which the tracer concentration is oscillates about some average value.

Case 1:

The monotonic increasing C(t), $C_{min} = C_1$ and equation (10) can be written as

$$\cdot \quad \lambda_{av}(t_1,t_2) - \frac{s_0}{\rho_{min}V_{min}C_1} \leq -\frac{1}{t_2-t_1}\ln(1+e_2) - \frac{s_0}{\rho_{min}V_{min}C_1}\frac{e_2}{1-e_2} \quad (14)$$

where $e_2 = (C_2 - C_1)/C_1$ is the relative increase in tracer concentration.

This expression allows one to estimate the error in the average air change rate when equilibrium have not been reached.

Case 2:

In the case of cyclic oscillations in the tracer concentration around some average value (This occurs when equilibrium has been reached, but the air change rate is not constant, but oscillates about some average value), one can choose t_2 such that $C_2 = C_1$ in which case

$$\frac{s_0}{\rho_{max}V_{max}C_{max}} \leq \lambda_{av}(t_1,t_2) \leq \frac{s_0}{\rho_{min}V_{min}C_{min}} \quad (15)$$

Application to Constant Concentration

For constant concentration, one attempts to control the injection of tracer such that a target concentration is obtained and held constant. In such an experiment one can in actuality maintain this target concentration only to within some tolerance. In practice the actual concentration in the zone oscillates about the target concentration and in this case one can choose t_2 such that $C_2 = C_1$. Equation (13) reduces to

$$\frac{S_{av}(t_1, t_2)}{\rho_{max} V_{max} C_{max}} \leq \lambda_{av}(t_1, t_2) \leq \frac{S_{av}(t_2, t_1)}{\rho_{min} V_{min} C_{min}} \tag{16}$$

Equation (13) can be used to bound the error in estimating the average air change rate if the control of the constant concentration system is not perfect.
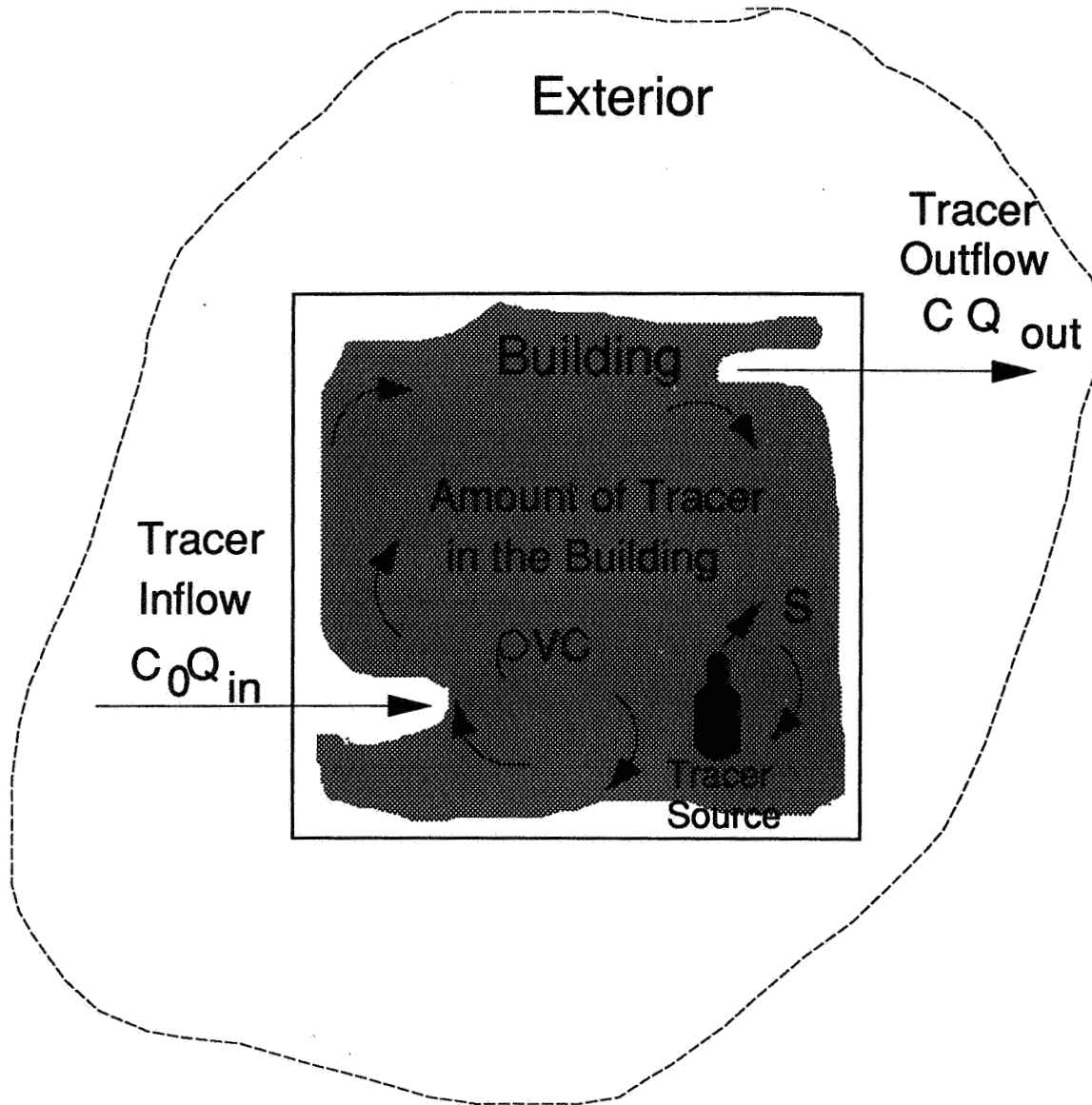
Single Zone Model

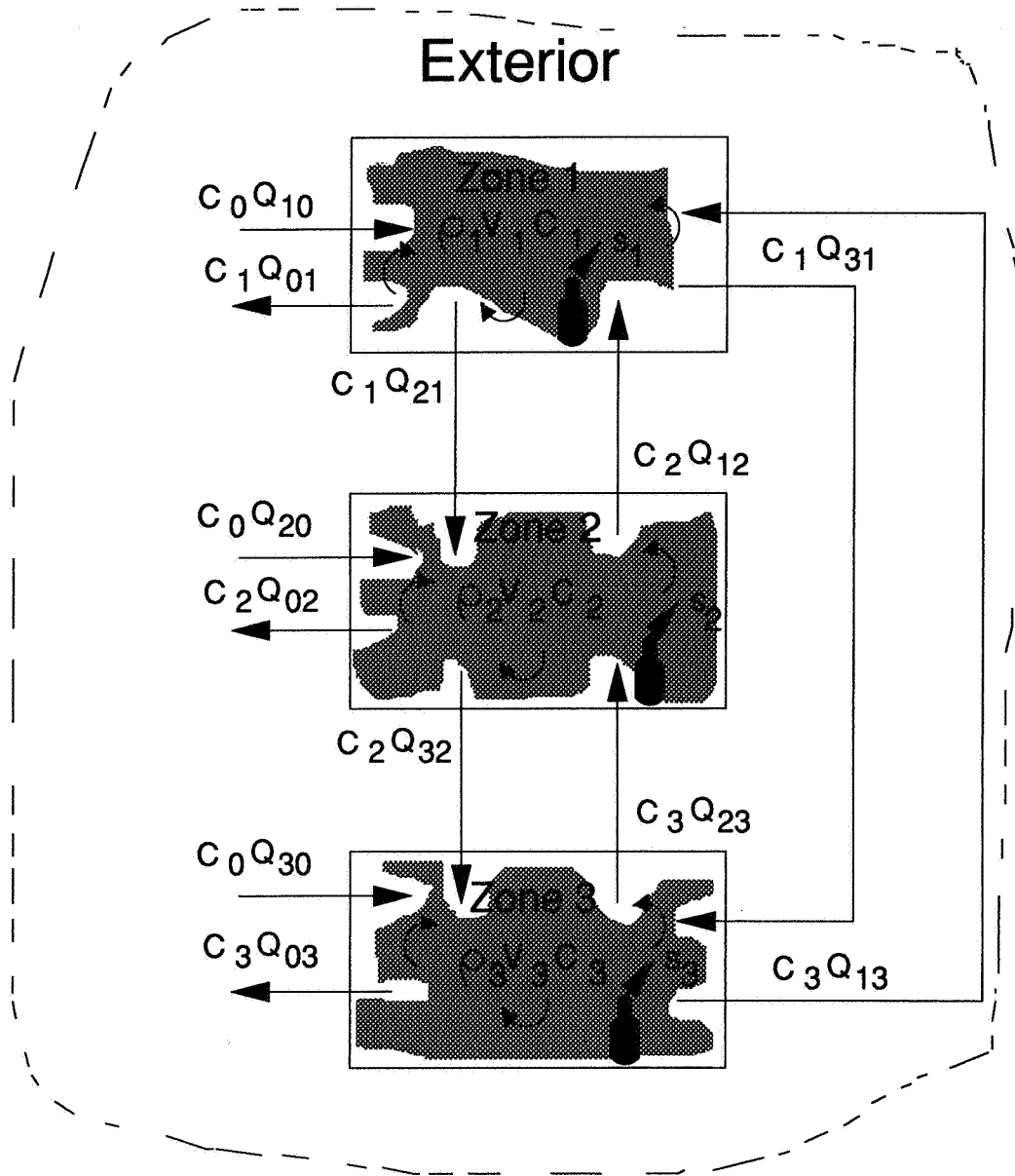Figure I-1. Single Zone Model for Tracer Experiments

Multi-Zone Model

Figure I-2. Multi Zone Model for Multi-Chamber Test