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VI MONITORING AND MODELING OF INDOOR AIR POLLUTION

This chapter discusses the research tools required for measuring or estimating indoor pollution and exposure to it.

Techniques and instruments used for the measurement of outdoor pollution may be modified for the sampling of indoor environments. Several problems emerge with such modifications, and those problems are discussed here, as well as instruments designed specifically for the sampling of indoor air.

Personal monitors are increasingly recognized as powerful scientific tools for determining individual and population exposure to air pollutants. Although they are still in the early stages of application, it is clear the personal monitors can yield data that are useful in associating human activities with exposure to air pollution. The benefits and deficiencies of personal monitors are discussed in a separate section of this chapter.

The extent of indoor air pollution can be estimated with numerical models; mass-balance equations are used to estimate concentrations of indoor pollutants as fractions of outdoor concentrations and to estimate infiltration rates, indoor source strengths, pollutant decay rates, and mixing factors. Several models have been developed, but few have been validated against data obtained from measurements.

In estimating the total exposure of humans to pollutants (exposure to pollutants encountered indoor and outdoors, in industrial sites and other workplaces, etc.), it is essential to know not only the pollutant concentrations, but also individual patterns of mobility and use of time. The available information pertinent to the last two characteristics has been gathered mostly by social scientists and, although interesting, does not meet the information needs for assessing exposure to air pollution. The final section of this chapter discusses the idea of total exposure and what knowledge is needed to measure it.

FIXED-STATION SAMPLING AND MONITORING

There is an extensive data base on outdoor air quality, and much of the knowledge gained from studies of outdoor air quality is applicable to the characterization of nonindustrial indoor environments. However, the characteristics of indoor air quality in residential and commercial buildings and at other indoor sites can be quite different from those of outdoor and heavy-industrial environments. Thus, a number of special problems arise: the quality of indoor air is affected by a broad spectrum of pollutants from both outdoor and indoor sources; measurements of indoor air concentrations may require sampling instruments considerably different from those used in the outdoor or industrial environment; and the air volume inside a building is finite, and the rate of air exchange (especially in residential units) may be very low, and therefore, when air samples are drawn from an occupied space by external samplers, the sampling flow rate must be so low as to have only a negligible effect on indoor air movement and on the air-exchange rate.

Because of the effects of equipment heat and noise, as well as occupant inconvenience, sampling and monitoring equipment should (and usually can) be placed in remote locations outside the building being evaluated. Thus, it is common practice is to locate the instruments outside the building space and draw air-sample streams to them.

Sampling techniques fall into the following broad categories:

- <u>Continuous sampling</u>; Provides "real-time" sampling; required to observe temporal fluctuations in concentration over short periods.
- <u>Integrated or continuous sampling</u>; Provides an average sampling over a specified period; used when the mean concentration is either desirable or adequate for the purpose.
- <u>Grab or spot sampling</u>: Provides single samples taken at specified intervals; typically consists of admitting an air sample into a previously evacuated vessel, drawing a sample into a deflated bag for later analysis, or drawing (by mechanical pump) a sample through a sample collector to extract a contaminant from the air; suitable when "spot" samples are adequate for the measurement of a pollutant and knowledge of temporal concentration variation over short periods is not important.

Some instruments sample and measure pollutants directly, and others sample for later laboratory analysis. The direct-reading instruments required for continuous monitoring use various types of physicochemical detectors that can measure the concentrations of pollutants in situ. Integrated or grab-sampling methods are used when there is no suitable concentration sensor available, when the pollutants of interest are present at concentrations too low to permit use of direct-reading instruments, or when sampling sites are inaccessible to bulky instruments. Further information on sampling and measurement methods for air pollutants is available elsewhere. ² ⁵³ ¹⁰³

CONTINUOUS MONITORING

Continuous monitoring is a technique for sampling and measuring the real-time concentration of pollutants. Indoor air quality is subject to both temporal and spatial variations, and data on these variations would be needed to determine the concentrations to which occupants are exposed or to model indoor air pollution. The choice of monitoring techniques must be consistent with the types of information desired and the resources and manpower available. Although continuous monitoring has numerous benefits, it also has a number of disadvantages.

Two positive features of continuous monitoring are that peak short-term concentrations can be determined, in addition to average concentrations calculated over any period, and that concentration variation as a function of time can be correlated with source generation, infiltration-ventilation, and other characteristics.

The availability of continuous-monitoring instrumentation depends on many factors, including the chemical properties of the pollutant and the range of concentrations to be measured. Continuous monitors are commercially available for all the gaseous pollutants that are designated "criteria" pollutants by the EPA—carbon monoxide, sulfur dioxide, nitrogen dioxide, ozone, and total nonmethane hydrocarbons. The EPA has specified performance criteria for the instruments used to measure each of these pollutants, and all analyzers that meet these specifications in performance sts are designated "EPA-approved."

Continuous-monitoring systems, even with high-quality instrumentation, are not trouble-free. For example, continuous monitors are expensive and require frequent calibration and routine maintenance. In addition, they have their own power and ambient-temperature requirements and can create safety, heat, and noise problems if they are placed at the sampling points. For these reasons, monitoring systems are generally designed to have all equipment for continuous analysis and recording at a single remote site, often a mobile laboratory. Such a laboratory usually contains facilities for calibration and maintenance, and it may also provide electric power and suitable environmental conditions for the equipment. If sampling lines made of flexible fluorocarbon tubing or other nonreactive materials are used, air from several sampling points can be drawn into the laboratory for analysis. One set of continuous monitoring equipment can be shared by several sampling sites if the individual lines are sequentially sampled. In this scheme, all instruments

obtain air samples from a common manifold, which, in turn, is supplied with air from one of four sampling sites (one of which is usually outdoors) or from a calibration system. The length of the sampling interval for each site can be determined by the response times of the individual instruments, the actual transit time in the sampling line, and the details of temporal information required at each sampling site.

Continuous monitoring requires highly trained field personnel, rigorous quality-control (calibration) procedures, and provisions for quality assurance (independent performance audits of routine monitoring and data-handling operations). Securing electric power and a suitable location for a mobile laboratory equipped with sampling lines and cables can require long-term planning and entail considerable expense. This type of fixed-station monitoring is not suitable for large-scale surveys, because of these time and cost considerations. For large-scale survey work, integrated sampling and grab-sampling techniques are generally more appropriate.

INTEGRATED SAMPLING

Integrated sampling, in which a known sampling rate is maintained over some period, is commonly used for pollutants that must be accumulated to permit analysis. The period of collection may be several minutes or several weeks or months. Analysis may be performed at the collection site or in a laboratory. The data resulting from analysis of integrated samples are expressed as an average concentration over the sampling period. A variety of particles, gases, and vapors are sampled by this technique.

Particles can be collected on filter media for later gravimetric and chemical analyses. Size-selective particle samplers, such as various dichotomous air samplers. and portable cyclone samplers, $\frac{10\ 102}{2}$ are used in indoor aerosol sampling when it is desirable to determine the concentration of fine particles (less than 2.5 μ m in aerodynamic diameter) or respirable particles (less than 3.5 μ m in aerodynamic diameter). The samples can be analyzed by beta gauge or gravimetric techniques capable of determining mass concentration; by x-ray fluorescence, neutron-activation analysis, etc., to determine their elemental composition; and by a variety of separation and analytic techniques to determine chemical composition. Aerosol samplers must be placed directly at the sampling sites, to avoid the particle losses that occur when air is drawn through sampling lines. The sophistication of particle samplers ranges from hand-held units that require manual operation to fully automated units that can be programed to operate unattended for several weeks.

Gaseous substances can be collected by both passive (diffusion-controlled) and active (powered bulk air-flow) samplers. Soluble vapors, such as formaldehyde and ammonia, can be collected by liquid gas washers and bubblers. Air sampling with bubblers, as well as with other accumulating sample collectors (such as adsorbers and condensation traps), requires that the total volume of the air sample be accurately known. This can be accomplished with dry- or wet-test meters, which measure sampled volume directly, or by measuring or controlling the sampling rate and time.

Many techniques have been used to measure the concentrations of radon and radon daughters. Because of the low level of radioactivity usually found in buildings, integrated measurements are often necessary. Passive devices that use sensitive thermoluminescent dosimeter (TLD) chips, passive film, or track-etch techniques can record alpha decay over periods of weeks or months to determine average radon concentrations. Radon-daughter concentrations can be determined by passing a known volume of air through a filter paper (typically for 10 min) and then measuring total alpha activity on the filter with an alpha-decay ratemeter.

Integrated sampling techniques have several advantages: they are less expensive and require less manpower than continuous monitors, they can be used to measure concentrations that are too low to be measured directly, samples can often be analyzed later at a more convenient time or place, and average concentrations over long periods are easily obtained. But they also have some disadvantages:

short-term temporal information is lost; samples must be taken frequently if temporal variability in concentration is to be assessed; transporting the sample to its point of analysis may require special handling, special environmental conditions, or rapid delivery to avoid deterioration; and quality control may be more difficult to implement.

GRAB SAMPLING

In grab sampling, one sample is collected over a very short period. Grab samples have to be taken frequently, if temporal variability in pollutant concentrations is to be assessed. It is usually the least expensive technique for field sampling, unless very frequent samples are required. It may simply involve filling a container with an air sample and transporting it to a laboratory for analysis, or it may involve extractive sampling, as in colorimetric detector tubes. It is most useful when the laboratory equipment required for analysis is at a remote location, when a very large number of samples are required, or when manpower and equipment are limited. Sampling vessels commonly used include plastic bottles, glass tubes filled with adsorbent, stainless-steel containers, and bags of aluminum polyester (Mylar), PVC film, and fluoroplastic film. 93 Grab sampling has been used to estimate concentrations of radon, tracer gases, and organic compounds.

Grab sampling can be used to measure radon concentrations by pumping a known volume of air through a filter into a Tedlar bag, which is impervious to radon. The time at which the sample is taken must be recorded, and, because of the decay properties of radon gas, analysis must be performed within a few days. If necessary, the sample can be concentrated with a cryogenic trap or transferred directly into a zinc sulfide scintillation chamber for alpha-counting. The bags are inexpensive and can be mailed, with manual pumps, to field sites. Similarly, air samples can be qualitatively and quantitatively analyzed for organic compounds with gas-chromatographic techniques.

Grab sampling, with its low cost and minimal manpower requirement, is suitable for large-scale survey work. However, a number of problems are associated with this technique. No information other than an "instantaneous" concentration can be obtained, and this value could be greatly affected by something as simple as the opening of a door or window. Sampled volumes are relatively small, and the laboratory measurement technique must be sensitive enough to determine ambient concentrations directly. Inward and outward diffusion of various gases has been observed for many materials used in collection bags, and leaks in the containers and connectors are common. Particular attention must be given to degradation, adsorption, contamination, transformation, and the possible formation of artifact pollutants. The expeditious transport of grab samples with reference to time, temperature, sealing, and handling is important. Quality control is difficult to maintain, but must be established before this technique can be used with confidence.

MONITORING OF VENTILATION RATE

Indoor air quality is directly affected by the rate at which outdoor air enters a building. Ventilation can be used to maintain low concentrations of indoor-generated pollutants. In turn, human comfort conditions, such as temperature and relative humidity, can be the determining factors in setting ventilation rates. Measurement of the infiltration-ventilation rate, the meteorologic factors that affect it (outdoor temperature, wind speed, and wind direction), and comfort factors (temperature and relative humidity) can be an integral part of fixed-location field monitoring.

Ventilation systems vary considerably. Detached single residential units are ventilated primarily by infiltration—the uncontrolled leakage of air through cracks in the building envelope (around doors and windows, through walls and floor joints, etc.)—and by the controlled opening of windows and doors. Large buildings are usually ventilated by mechanical systems of varied complexity.

So-called fresh air enters detached residential structures by infiltration; the term "air changes per hour" (ach) is routinely used for this source of ventilation—"1 ach" means that a volume of outdoor air equal to the volume of the interior building space "leaks" inside each hour. That does not imply that the incoming air drives out or displaces the old air as it enters; rather, it is assumed that perfect mixing takes place. In practice, however, perfect mixing is impossible to achieve. Therefore, an estimate of outdoor-air flow rate is based on the assumption of perfect mixing and homogeneity of indoor air to facilitate calculating infiltration rates.

By far the most commonly used method of estimating air-exchange rates is the tracer-gas decay technique. In this method, a tracer gas is released into the building space at one or more points, possibly with the use of fans. In this way, an attempt is made to produce a uniform concentration throughout the building space. If homogeneity is maintained, the decay of the tracer gas is exponential, and the infiltration rates can be determined by sampling the air at several times. The air-exchange rate can be obtained from the slope of a semilogarithmic plot of the natural logarithms of the pollutant concentration versus time.

In a similar method, the equilibrium-concentration method, a tracer gas is released at a constant rate into the building space. In the steady-state condition with perfect mixing, the indoor concentration will reach a steady-state value. From this and the injection rate, the infiltration rate can be calculated. With this technique, although it is simple to perform, it often takes many hours to reach a steady-state equilibrium.

More complex tracer-gas systems can measure infiltration rates on a semicontinuous or continuous basis. Many gases have been used for tracer-gas measurements. Some of the properties that such a gas should have are easy measurement at low concentrations, minimal interference from other air constituents, chemical stability, nonreactivity, lack of absorption by building contents, a density comparable with that of air, safety for humans, lack of explosiveness and flammability, absence of other interior or exterior sources, low cost, and ready availability. Some gases commonly used are sulfur hexafluoride, nitrous oxide, and ethane. 40

Mechanical ventilation systems vary considerably in design and complexity, and methods chosen to estimate ventilation rates must be suitable for the systems under consideration. The methods commonly used to estimate the ventilation rate for systems that use recirculation include pressure-measuring devices (such as inclined manometers and U-tubes), velocity meters (such as pitot tubes, hot-wire flowmeters, heated-thermistor flowmeters, and heated-thermocouple flowmeters), mechanical gas-flow indicators (such as rotating and deflecting-vane anemometers), tracer-gas techniques, and heat-balance techniques. Care must be taken to distinguish between the total rate at which air enters a particular zone and the rate at which outside air enters the zone.

Temperature-measuring devices suitable for continuous monitoring include thermocouples, semiconductors, and thermistors. Typical indoor temperatures range from 15 to 40°C. Thermocouples present problems with low voltage outputs near 0°C and have nonlinear characteristics, but only when the cold junction is at 0°C. Semiconductor temperature sensors that use integrated circuits and have a voltage output linear with temperature are suitable for continuous recording. The most common temperature probe for measuring temperatures in this range is probably the thermistor, because of its high resistance ratio (which yields large voltage changes for small changes in temperature), its linearized output, and its wide operating range. Temperature gradients can be large in a building and even in an individual room. Probes should be placed where they will sense the temperature experienced by the occupants. Temperature probes should be calibrated against mercury thermometers that meet the specifications of the American Society for Testing and Materials (ASTM).

Relative humidity can be measured with sensors of the "human hair" type, which expand and contract with changes in humidity, or with dewpoint-measuring devices. Commercially available dewpoint hygrometers, based on the principle that the vapor pressure of water is decreased by the pressure of an inorganic salt, are well suited to continuous monitoring. Relative humidity can be readily calculated from dry-bulb and dewpoint temperatures. Relative-humidity measuring devices can be calibrated with the aid of sling psychrometers.

PERSONAL MONITORS

Over the last 2 decades, a wide variety of miniaturized air samplers have become available that collect gaseous and particulate samples from the immediate vicinity of people, even as they conduct their normal activities. The initial devices used battery-powered samplers, defined as "nonpassive." Although widely used, these devices are often larger and heavier than desirable. More recently, a variety of diffusion- and permeation-controlled samplers have become available. These "passive" devices are applicable solely to gas- and vapor-sampling and are very snail and light. They all use sensitive chemical or physical analytic methods.

Three recent workshops reviewed candidate technologies for personal sampling and monitoring of air-pollution exposure. A Brookhaven group identified potential methods for gas- and particle-monitoring. An EPA feasibility study identified useful methods for monitoring sulfur dioxide, nitrogen dioxide, and ozone. Another EPA symposium explored the use of available technology for health-effects studies and other uses.

Blood carboxyhemoglobin (COHb) can be used as a measure of the actual dose of carbon monoxide received by a person. Respirable-particle concentrations are also of prime concern in health-effects studies; some of the factors involved in obtaining reliable data have been evaluated. 91 102

PERSONAL SAMPLING DEVICES

Gas-Sampling

The major techniques developed for sampling gaseous pollutants are passive (based on membrane permeation or diffusion through a geometrically defined air space) and nonpassive (in which air-pumping devices draw defined air volumes through devices of known collection efficiency).

<u>Passive Samplers</u>. Passive samplers use the kinetic energy of gas molecules and the efficiency of the adsorbent collector to extract pollutant molecules from the air at a known rate. The sampler must be placed at the collection site, but has no requirement for a pump, flow regulator, or batteries. Such samplers therefore have major advantages with respect to weight, cost, and maintenance. There are two basic types: diffusion and permeation. Their use is limited by the rate and amount of gaseous diffusion through a geometrically controlled air space or by transport through a permeable membrane that is specific for the pollutants being sampled.

The choice and use of the diffusion-collector technique require knowledge of the coefficient of diffusion of the pollutant to be sampled in air under conditions similar to those normally encountered. Humidity effects have been encountered; these are most probably caused by changing absorbent efficiency.

One diffusion sampler has been developed 63–85 for the measurement of ambient nitrogen dioxide. It uses the principle of diffusion through the bore of an open tube that defines the rate of transport to the collector. The quantity of nitrogen dioxide diffused from the open end of the tube to the collector surface (triethanolamine) is calculable by Fick's first law of diffusion, which may be expressed as:

=D(A/L)Ct, where =number of moles of nitrogen dioxide transferred during time t, A=cross-sectional area of tube (cm²), L=distance from open end to collector surface (cm), C=concentration difference between tube entrance and closed end (mol/cm³), and D= coefficient of diffusion of nitrogen dioxide in air (cm²/s). The required nitrogen dioxide absorbed is given as a time-weighted average concentration for the sampling period.

Substituting typical values of the parameters—D=0.154 cm²/s, A=0.71 cm², and L=7.1 cm—simplifies

the expression to: $^{\mathbf{Q}}$ =2.3 (ppm-h)×10⁻⁹.

In principle, this method is applicable to determination of any gaseous air pollutant for which an efficient, selective absorbent is available and for which an appropriate analytic chemical procedure may be devised. The size of the sampler can be varied, but attention must be given to scaling factors. All 80 Diffusion samplers have reportedly been used for water vapor and sulfur dioxide, an itric oxide, and sulfur dioxide, ammonia, ammonia, ammonia, ammonia, and and NO_x. An activated-carbon element has been used as the collector in a badge that has an open grid to define the geometry of the gaseous diffusion port. This method of sampling requires use of gas-chromatographic (GC) analysis for measurement of the specific gaseous pollutant absorbed. A variety of organic compounds can be sampled and measured by this technique.

A large variety of permeation samplers used for monitor systems are available commercially. All use membranes fabricated and calibrated to control the rate of permeation of the pollutant to the collector, which may be a solid medium, such as charcoal or Tenax GC for specific chemicals. Processing of the collected sample varies widely; chromatographic or colorimetric procedures are commonly applied for measurement.

Transport of a gaseous pollutant across a membrane resembles the diffusion process. However, permeation involves solution of the gaseous species in the membrane. Specific interaction between the gas and the polymer matrix introduces variables. As in a diffusion collector, the concentration of the gas approaches zero on the side of the membrane next to the collector, causing a gradient that results in flow from the ambient-air side.

The permeability constant, P, of a membrane is defined by the equation N=PA(C_1 – C_2)/S, where N=rate of transport across the membrane (mol/s), P=permeability constant (cm²/s), A=cross-sectional area (cm²), S=membrane thickness (cm), C_1 = concentration of gas on ambient side of membrane (μ g/m³), and C_2 =concentration of gas on collector side of membrane (μ g/m³). As a function of time, Nt=PAC₁t/S. If W=Nt, the amount of gas that passes through the membrane in time t, then, because P, A, and s are constant for any device and gas, W=C₁t/k, where k=s/PA, or C₁=Wk/t.

Thus, the amount of a gaseous pollutant, W, trapped in the collector medium is proportional to its concentration, C_1 , in the air. The value of k must be determined by calibration.

Membranes used in permeation collectors are made from polymeric materials, such as dimethylsilicones, silicone polycarbonate, silicones, cellulose acetate, TFE Teflon, FEP Teflon, Mylar, polyvinyl fluoride, Iolon, and Silastic. Thicknesses of $2.5-25\times10^{-3}$ cm have been tested.⁸⁸

The permeation collector with activated absorbents is particularly useful for organic pollutants when GC analysis is applied. Commercial monitors are available from the 3M Corporation 35 and Du Pont that allow determination of more than 80 compounds by this method. Sulfur dioxide, 63 68 107

chlorine, $\underline{^{37\ 107}}$ vinyl chloride, $\underline{^{75\ 105\ 107}}$ nitrogen dioxide, $\underline{^{106}}$ alkyl lead, $\underline{^{107}}$ and benzene $\underline{^{107}}$ have been determined. The utility of a spectrum of absorbents in these passive collectors has been tested. 9

<u>Nonpassive Systems</u>. A considerable variety of sampling systems using pumps to move the air have come into use over the last 2 decades, including impinger systems and solid adsorbers for gases and impingers, filters, and impactors for solid particles. Directly indicating devices using impregnated papers, chalks, and crayons and stain-detector tubes have also been used. These techniques have been reviewed in detail by Linch⁵⁵ and Saltzman.⁹¹

A recent development in personal monitors involves a pump that is positioned next to the wearer's diaphragm by a light harness. The volume of air pumped by the motion of the thoracic cavity is recorded by an electronic package, which may be checked by a detached readout system. This system samples air for gases through coated diffusion-tube collectors or for particles through small filters at flow rates of 75–500 ml/min, depending on the wearer's breathing rate. The complete apparatus weighs 590 g. In conjunction with spirometer calibration, actual exposures to measured pollutants may be calculated.

Particle-Sampling

Particulate samples are collected by using the same principles used for large-scale samplers: filtration, impaction, and liquid impingement. The separation of the respirable fraction of particles is of considerable importance for personal monitors, and collection of adequate numbers of samples for analysis is critical. All particle-samplers use some device for moving the air sample and for separating the respirable-particle fraction. Collection is preceded by such a device as a cyclone presampler. The relatively low power available to drive the air-sampling pump usually limits particle collection by personal monitors to filtration, either in a single stage or in a second stage that follows a cyclone that collects the large, nonrespirable particles. For respirable-particle-sampling, the air-flow rate must be precisely controlled.

The theory of aerosol collection by filtration has been extensively reviewed by Dorman, ²¹ Pich, ⁸⁷ Fuchs, ²⁶ Green and Lane, ³⁶ Liu and Lee, ⁵⁷ and Lippmann. ⁵⁶ Small-scale impactors suitable for respirable-dust-sampling with a personal monitor have been described by Marple ⁶¹ and Willeke. ¹⁰⁸

Biologic Monitoring

Measurements made on the human body and its excretions constitute an alternative way of measuring exposure to environmental pollutants. They include measurements of blood, urine, feces, and hair. The methods that can be used to relate environmental pollutant exposure to human composition have been reviewed at length in other NRC reports (on carbon monoxide, nitrogen oxides, and various trace metals).

USE OF PERSONAL MONITORS IN EXPOSURE STUDIES

Exposures to air pollution usually vary with a person's mobility patterns and activities. Therefore, estimating the total exposure of a person from one or a few air-pollution measurements at stationary locations cannot properly characterize the variation in a population's or a person's actual exposures. To evaluate health effects, it is necessary to know actual personal exposures and the distribution of those exposures in a population. The need for direct measurement of personal exposure to pollution has been noted by several authors. 49 67

Personal monitors for various pollutants are commercially available. The Brookhaven workshop identified four basic experimental designs:⁶⁸

1. <u>Use of Individual Air Pollution Monitors for Direct Determination of Exposure</u>. Each person in the study population would wear or carry an individual air pollution monitor during the course of the

study. The same individuals would also be subjected to continuous or periodic evaluation of health responses. Individual exposure and response would thus be measured.... Because of economic constraints, only relatively small populations could be studied by this direct approach.

- 2. <u>Use of Individual Air Pollution Monitors to Adjust Results from Fixed Stations</u>. As previously indicated, there can be substantial variations between area level measurements and personal exposure measurements. By monitoring exposure of individuals with individual air pollution monitors in areas also monitored with fixed stations, one would obtain the distribution of individual exposures in relation to measurements obtained at the fixed stations. If one or several relatively constant relations were found in various areas, fixed-station data would then be corrected for use in estimating population exposures.
- 3. <u>Use of Representative Sampling to Determine Subgroup Exposure</u>. A carefully selected sample of the study population would be asked to wear or carry individual air pollution monitors. The sample would be stratified, grouping those expected to have similar exposures (e.g., office workers or street workers). The measured exposure of each subgroup in the sample could be used as representative of the entire group.
- 4. <u>Use of Individual Air Pollution Monitors to Calibrate Personal Activity Models</u>. Activity models have been developed that describe how and where people spend their time.... These models could prove to be useful in estimating population exposure. They have not been applied in air pollution epidemiology except in a very limited way, and they could be best calibrated or verified through experiments using individual air pollution monitors. In such an experiment, a carefully selected sample of the study population would be asked to wear individual air pollution monitors, and their measured exposure would be compared with the estimated exposure of the activity model.

Gaseous Pollutants

<u>Carbon Monoxide</u>. Ott and Mage⁷⁹ collected 425 integrated carbon monoxide samples over 21 d in November 1970 and January 1971 in downtown San Jose, California. Breathing-zone samples collected while the subjects walked typical pedestrian routes were compared with those measured at the fixed monitoring stations. The mean pedestrian exposure was 1.6 times the mean concentration measured by the fixed monitors, but individual measurements of exposure varied from those measured at the fixed stations by a factor of up to 10. Ott and Mage concluded that the fixed-station monitoring data "provide a relatively poor measure of the true exposure of members of the general public to air pollutants."

Wright <u>et al</u>.¹¹¹ sampled exposures in Toronto with portable carbon monoxide monitors. They demonstrated a "substantial discrepancy between the carbon monoxide concentrations detected by the provincial network of fixed-site sampling stations and the much higher concentrations commonly met by people living and working in a large metropolitan area such as Toronto."

Cortese and Spengler ___ measured exposure of Boston commuters equipped with portable carbon monoxide instruments (Ecolyzers). They reported that 1-h exposures exceeded fixed-monitor measurements by a factor of 1.3–2.1; 8-h mean exposures were considerably below the 8-h mean from the fixed monitor.

Wallace 104 carried a carbon monoxide dosimeter during 30 commuting trips by bus to his office in Washington, D.C. Concentrations inside the vehicles were typically 2–4 times those continuously measured in the central city at the fixed monitoring station (Figure VI-1). There was no correlation between the ambient and personal in-vehicle measurements.

<u>Nitrogen Dioxide</u>. In a personal-monitoring study of children in Ansonia, Connecticut, ⁶ nitrogen dioxide and sulfur dioxide were measured with bubblers. ¹⁰ Twenty boys were equipped with

suitcase samplers that they carried for one 24-h day. Exposure to nitrogen dioxide tended to be greater in children exposed to smoking at home, but the differences were not statistically significant. As with sulfur dioxide, mean personal exposure values (61.3 \pm 7.2 μ g/m³) were significantly lower than mean outdoor nitrogen dioxide concentrations (100.1 \pm 9.0 μ g/m³).

Palmes <u>et al</u>. ⁸⁴ has described a passive personal sampler for nitrogen dioxide suitable for occupational exposures. The same device has been used to estimate 1-wk average indoor nitrogen dioxide concentrations in 109 dwellings with gas stoves and nine with electric stoves in metropolitan New York. ⁸⁶ It was found that the homes with gas stoves had significantly higher nitrogen dioxide concentrations than those with electric stoves. Average values in the kitchens with gas stoves approached the U.S. primary ambient-air quality standard of 50 ppb (annual average).

These dosimeters were used in a personal-monitoring study of five families with gas stoves and four families with electric stoves in Topeka, Kansas. In each family, the husband, wife, and one child wore the dosimeters for four 1-wk samples. Dosimeters were also placed outside, in the kitchen, and in the bedroom. No significant differences were found between the personal-monitor and outdoor measurements for the families with electric stoves (Figure VI-2). For the families with gas stoves, personal exposures were significantly higher than outdoor values and correlated best with the fixed dosimeters in the bedroom. No significant differences in exposures were found between family members.

<u>Sulfur Dioxide</u>. Exposure to sulfur dioxide has been estimated in several studies by the calculation of a time-weighted average exposure from the time spent and average concentrations in various places. Sulfur dioxide personal monitors have not been extensively used in field studies.

In the personal-monitoring study of children in Ansonia, Connecticut, no significant difference was found among the sulfur dioxide personal exposure measurements of the boys. The personal samplers had a mean of 5.5±0.07 μ g/m³, which was significantly lower than the outdoor mean of 12.0±2.2 μ g/m³. The reports of daily activities showed that the children were indoors between 60% and 80% of the day.

Passive personal monitors using the collection principle of gas permeation through polymer membranes have been shown to be sensitive to 24-h average concentrations of sulfur dioxide down to 0.01 ppm. Sampling at ambient concentrations with treated filters also appears feasible. However, no personal-monitoring results with these techniques have been reported.

<u>Organic Substances</u>. Several passive monitors have been developed that may have the sensitivity to measure mean personal exposure to organic substances for sampling periods of 1 d to 1 wk. $\frac{9 \ 32 \ 33 \ 35}{2}$ Their use has yet to be demonstrated in personal-monitoring programs in nonindustrial environments.

Particles

Respirable Particles. In the Ansonia study, 6 personal exposures to respirable particles were significantly higher among children who lived with one or more smokers. The mean personal exposure, $114.5 \pm 9.0 \, \mu g/m^3$, was significantly higher than the mean outdoor concentration, $58.4 \pm 5.9 \, \mu g/m^3$. The outdoor high-volume samplers, however, collected both respirable and nonrespirable particles. The principal conclusion of this study was that a child's exposure "load" of air pollutants, especially respirable particles, is determined primarily by indoor exposures.

Personal exposure to respirable particles and sulfates has been measured in two cities as part of the Harvard six-city study, in which 37 people carried personal respirable-particle monitors with them during the day. Fixed-station monitors were run simultaneously in the main activity room of each

home and at several locations outside. There were at least three complete sample days for each person.

Mean personal exposures to respirable particles and sulfates for each city were determined on the basis of mean outdoor concentrations. In each city, there were significant differences in results between individuals, as determined by their activities. A linear increase in personal exposure to respirable particles with the number of smokers in the home and workplace was found.

Lead. Berlandi et al. have reported personal lead-exposure measurements from 2 d of sampling in metropolitan Boston. Samples collected while subjects were driving into Boston had a time-weighted average of 4.5 μ g/m³ the first day and 3.7 μ g/m³ the second; indoor personal samples were all less than 1 μ g/m³. Fugaš et al. estimated average air lead exposure of an office worker in Zagreb. Air lead was measured with a personal monitor inside and outside her home and her office and at other sites. Table VI-1 shows that the average concentrations were highest in association with outdoor activities—6.3 μ g/m³—with only 1.7 μ g/m³ or less from indoor sampling. A time-weighted average exposure was then calculated on the basis of her activities each week (Table VI-1). The average weekly exposure was estimated to be 1.1 μ g/m³, compared with the average measured value of 0.72 μ g/m³ outside the subject's home.

Fugaš 27 extended this method of estimating exposure to lead in air to a middle-sized industrial town in Yugoslavia. Air lead was measured at various locations indoors and outdoors during the winter of 1972–1973. Estimated exposures were considerably higher than the average urban monitoring-station value of 0.9 μ g/m 3 .

Lead concentrations up to 10 times that found in buildings may be found in vehicles or outdoors. The contribution of outdoor and vehicular exposure to mean personal exposures may be small, because of the comparatively short exposure times.

Biologic Indicators

Direct measurement of individual dose by biologic means is possible for several pollutants. This method intrinsically compensates for different rates of uptake by different persons, as well as for differences in exposure.

<u>Carbon Monoxide</u>. Actual carbon monoxide dose received can be measured directly by measuring blood carboxyhemoglobin.

Stewart et al. 99 100 measured COHb of blood donors in 26 American cities. They reported COHb concentrations higher than those expected from fixed-station monitoring data. Goldmuntz compared COHb measurements for nonsmokers from 1969 through 1972 with carbon monoxide measurements from 37 EPA fixed-station monitors in 1973. Goldmuntz argued that the measurements from some of these fixed stations may be inappropriately high, because of their siting.

Morgan and Morris⁶⁸ calculated COHb concentrations that would be expected if the population were in equilibrium with the measured fixed-station carbon monoxide concentrations by the relation, % COHb= 0.16[CO]+0.3, where [CO] is the carbon monoxide concentration (ppm). Comparing these calculations with nonsmoker COHb measurements, they concluded that "the average dose indicated by COHb levels exceeded that predicted from the fixed-station data by a factor of 2."

Stewart <u>et al.</u>,¹⁰⁰ in a similar analysis of data obtained in Chicago in 1970 and 1974, reported COHb concentrations close to, but consistently higher than, those predicted from fixed-station measurements. They noted that the use of fixed-station measurements to define population exposure may not reflect worst-exposure situations, indoor exposures to carbon monoxide from cigarette smoke, or exposures from faulty heating systems. Kahn <u>et al</u>.⁵⁰ demonstrated that COHb

concentrations among nonsmokers in the St. Louis population are strongly affected by occupational exposure and by exposure to smokers.

<u>Lead</u>. There is little doubt of a correlation between the high exposures to lead in the air of industrial areas and indexes of lead absorption, such as blood lead, urinary lead, delta-aminolevulinic acid dehydrase (ALAD), and delta-aminolevulinic acid (DALA). The relationship between these measures of lead dose and the lower concentrations of air lead characteristic of the community—i.e., less than $10 \, \mu \text{g/m}^3$ —is not well established. A National Research Council committee has stated that "more precise studies are needed of the relation between atmospheric lead exposure in the urban environment and the concentration of lead in the blood, perhaps by the use of personal monitors."

In an attempt to characterize this relationship, Azar <u>et al</u>.³ compared average exposures to lead in air measured by personal monitoring with the biologic indexes of lead absorption. Two groups of 30 taxi drivers in two cities and three groups of 30 Du Pont employees in three cities carried personal particle monitors with them for 2–4 wk. Their exposure to air lead was calculated as a time-weighted mean for their exposures at home and at work. Blood lead was determined weekly, and urinary lead daily. Different relationships were found between average exposure to airborne lead and the logarithm of the blood lead concentration in each city. The plots of the data for all five groups, however, had a similar slope, with different intercepts. The authors suggested that the different intercepts indicate that variables other than airborne lead, presumably ingested lead, are affecting blood lead content. No comparisons with fixed monitoring were made.

MODELING OF INDOOR AIR QUALITY

The value of an indoor air pollution model is twofold. First, it provides a framework for interpreting experimental results and for planning new experiments. Specifically, a model is useful in relating indoor pollutant concentrations to various geometric, ventilation, source, and sink parameters. Modeling can be used to determine the accuracy and precision to which various quantities must be measured if the desired accuracy of prediction is to be achieved. It can also be used in sorting out trends in the experimental data.

Second, and more important, a model provides a means to predict accurately some desired function of concentration (such as peak concentration or dosage) for places and conditions other than those tested experimentally.

In epidemiologic studies, it is important to consider the quality of the air to which subjects are actually exposed; in many cases, the air quality associated with the home, the mode of transportation, and the workplace should not be taken to be the same as that associated with the outside.

Indoor-air-quality models are developed to aid in understanding and predicting indoor air-pollutant concentrations and dosages as functions of outdoor air-pollutant concentrations, indoor-outdoor air-exchange rates, and indoor air-pollutant sources and sinks.

Air pollution indoors may be of outdoor or indoor origin. Outdoor pollutants may enter a structure through infiltration or ventilation. Pollutants of indoor origin may arise from point or diffuse sources. Regardless of their source, air pollutants may be transported and dispersed throughout various regions of the enclosure. Some pollutants may be removed by filters through which the makeup air or the recirculated air flows, by exfiltration or ventilation to the outdoors, and by chemical change. In the case of particles, surface removal and generation are often important.

Given familiarity with the system to be described and with the purpose of developing and using an indoor air pollution model, the starting point in developing a model is usually a statement of the mass balance concerning the pollutant of interest. For example, consider a structure of volume V, in which makeup air enters from the outside and passes through a filter at a rate q_0 . Part of the building air is recirculated through another filter at a rate q_1 , and air infiltrates the structure at a rate q_2 . Each filter

is characterized by a factor $F = (C_{inlet} - C_{outlet})/C_{inlet}$. In this example, the pollutant concentration is assumed to be uniform throughout the structure. The indoor and outdoor pollutant concentrations at time t are C and C_0 , respectively. The rate at which the pollutant is added to the indoor air owing to internal sources is S. The rate at which the pollutant is removed from the air owing to internal sinks is R. In this case, the appropriate starting equation is:

$$V \frac{dC}{dt} = q_0C_0 (1 - F_0) + q_1C (1 - F_1) + qC_0 - (Q_0 + q_1 + q_2)C + S - R.$$
Input rate due to makeup, recircu-
lated, and infiltrated air

Output rate

Source Sink
rate rate

(1)

The decay rate is a function of C; however, in modeling indoor air quality, the sink rate is often considered constant. As indicated in Equation 1, solutions to mass-balance equations invariably contain parameters that must be evaluated independently. Geometric parameters, such as volumes and surface areas, can be measured directly or obtained from blueprints. Accurate values of the ventilation parameters are usually more difficult to determine. Experimental techniques for obtaining net exchange rates between indoor and outdoor air have been reviewed by Georgii³⁰ and Gilath. The use of sulfur hexafluoride, SF₆, as a tracer for air-exchange-rate studies appears to be increasing; air samples can be collected by hand in the region of interest, remotely, or with automated instruments. Without forced ventilation, air exchange is due primarily to infiltration; in most forced-ventilation systems (which are balanced so that negative pressures are not created inside the building), the rate of infiltration is negligible, compared with the forced-ventilation rate.

Although ventilation parameters are often hard to obtain, the most difficult to evaluate are usually those associated with the rate at which the pollutant is being released or being removed (i.e., the strengths of the sources and sinks).

In view of the uncertainties associated with many of the parameter values and the difficulty of doing otherwise, compartments have been widely used in modeling indoor air quality. Traditionally, a compartment is defined as a region within which spatial variations in pollutant concentrations can be neglected over the time scale of interest. At any given instant, the concentration of a pollutant might vary substantially throughout the region of interest. However, if dosages are similar throughout the region of interest over intervals that are shorter than the time during which receptors (people, plants, equipment, etc.) are exposed, and if the damage is primarily a function of the dose, then the region may be treated as a compartment.

Depending on ventilation conditions, a single room, a floor, or a whole building may be adequately approximated as a single compartment. However, when either sources or sinks are not uniformly distributed throughout the region of interest, and the rate of mixing throughout the region of interest is low, compared with the characteristic residence time, then the single-compartment model may not provide an adequate description. For example, stratification in a room cannot be neglected when one is describing the movement of smoke and toxic gases associated with building fires; however, even in such a case (where intense stratification is to be expected), only two compartments (coupled) were needed to obtain a satisfactory description.

Sulfur hexafluoride tracer experiments, conducted with average-sized rooms ($20 \times 20 \times 8$ ft) in which one or more persons were moving and in which the air was being exchanged about 3 times per hour, have suggested that associated eddy diffusivities are around 10^3 cm²/s (D.D.Reible and F.H.Shair, personal communication); thus, about 5 min after an instantaneous point-source release, tracer concentrations (although decreasing) were about equal throughout most of a room.

The solution to the two-compartment model with constant coefficients is presented below, after a brief general discussion of multicompartment models. Examples of two-compartment and single-compartment models of indoor air quality are also discussed.

MULTICOMPARTMENT MODELS

Most n-compartment models have been (or probably will be) described by n coupled first-order linear ordinary differential equations of the form:

$$\frac{dx_1}{dt} + a_1x_1 = a_2x_2 + a_3x_3 + a_nx_n + a_n + 1$$

$$\frac{dx_2}{dt} + b_1x_2 = b_2x_1 + b_3x_3 + b_nx_n + b_n + 1$$

$$\frac{dx_n}{dt} + \cdots$$

In general, the terms a_1 , b_1 , etc., represent the sum of first-order losses from the compartment due to exhaust streams, filtration of any recirculating streams, and sources and sinks due to first-order chemical reactions. In most cases, the sources and sinks due to chemical reactions may be simulated as pseudo first-order, because of the low concentrations (parts per million, or less) of the pollutant. In cases where higher-order chemical reactions are important, the model equations will be nonlinear and generally will have to be solved numerically. In the case of particulate pollutants, the parameters a_1 , b_1 , etc., will probably contain loss terms, owing to surface deposition. The coefficients $a_2...a_n$, $b_2...$ b_n , etc., represent the gain of pollutants in various compartments that may result from the intrusion of air from other compartments. The terms a_{n+1} , b_{n+1} , etc., represent the sums of the zeroth-order source and sink terms associated with each of the compartments.

As indicated by Equations 2, an n-compartment model will contain n(n+1) parameters, whose values should be determined independently. Any temptation merely to fit the data through blind adjustment of the values of the parameters should be resisted, if the model is to be of broad value. The aim of any model should be to explain (and predict) as many data as possible with the smallest possible number of "adjustable parameters."

Because it is always possible to define x_n as the concentration of the pollutant in the nth compartment at any time minus the initial concentration, the initial conditions for Equations 2 may be taken as $=x_2(0)=...x_n(0)=0$.

TWO-COMPARTMENT MODELS

In general, the equations that describe two-compartment models are of the form:

$$\frac{dx_1}{dt} + a_1x_1 = a_2x_2 + a_3.$$

(3)

and

$$\frac{dx_2}{dt} + b_1x_2 = b_2x_1 + b_3.$$

(4)

The initial conditions are:

$$x_1=x_2=0$$
 at t=0. (5)

When the coefficients of Equations 3 and 4 are constant,

$$X_{1} = \frac{a_{3}b_{1} + a_{2}b_{3}}{a_{1}b_{1} - a_{2}b_{2}} + \left(\frac{a_{3}b_{1} + a_{2}b_{3}}{a_{1}b_{1} + a_{2}b_{2}}\right) \left(\frac{\beta e^{-\alpha t} - \alpha e^{-\beta t}}{\left[\left(a_{1} - b_{1}\right)^{2} + 4a_{2}b_{2}\right]^{\frac{\alpha}{2}}}\right) - a_{3} \left(\frac{e^{-\alpha t} - e^{-\beta t}}{\left[\left(a_{1} - b_{1}\right)^{2} + 4a_{2}b_{2}\right]^{\frac{\alpha}{2}}}\right)$$

(6)

and

$$X_{2} = \frac{a_{1}b_{3} + a_{3}b_{2}}{a_{1}b_{1} - a_{2}b_{2}} + \left(\frac{a_{1}b_{3} + a_{3}b_{2}}{a_{1}b_{1} - a_{2}b_{2}}\right) \left(\frac{\beta e^{-\alpha t} - \alpha e^{-\beta t}}{\left[\left(a_{1} - b_{1}\right)^{2} + 4a_{2}b_{2}\right]^{\frac{1}{2}}}\right) - b_{3} \left(\frac{e^{-\alpha t} - e^{-\beta t}}{\left[\left(a_{1} - b_{1}\right)^{2} + 4a_{2}b_{2}\right]^{\frac{1}{2}}}\right)$$

(7)

where

$$\alpha \equiv (a_1 + b_1)/2 + [(a_1 - b_1)^2 + 4a_2b_2]^{\frac{1}{2}}/2$$

(8)

and

$$\beta \equiv (a_1 + b_1)/2 - [(a_1 - b_1)^2 + 4a_2b_2]^{1/2}/2.$$

(9)

In Equations 6 and 7, the first term on the right side represents the steady-state solutions that are reached after the transient terms decay.

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Woods <u>et al</u>.¹¹⁰ used a two-compartment model in their analysis of thermal and ventilation requirements for laboratory-animal cage environments (see also Woods¹⁰⁹). The two compartments were the room and the animal cage. Mass or energy balances for each compartment were coupled by both free convection and forced circulation of room air through the cage. Their models permit estimation of dry-bulb and dew-point temperatures and concentrations of gaseous particulate contaminants in cages, as well as in a laboratory room. Such models can be used to determine an acceptable means of safely reducing room ventilation rates with implications of reduced energy consumption and operational costs.

Miller has used a two-compartment model in his description of the reentry of the exhausts from laboratory fume hoods. The two compartments were the building and the building wake (from which the makeup air is drawn). Miller had sasaki et al. have shown that the reentry of fume-hood exhaust is a much more pervasive problem than is commonly recognized. Sulfur hexafluoride tracer experiments have shown that reentry of a portion of the fume-hood exhaust is usually the dominant factor in determining the concentrations of the pollutants to which all persons are exposed in the laboratory building. Indoor concentrations of fume-hood exhausts, normalized to the source strengths, range from about 1 to about 350 ppb per mole released per hour. Although the chemical nature of the fume-hood emission is of prime importance, persons typically complain often when they are in buildings whose normalized indoor concentrations are above 100 ppb per mole released per hour from fume hoods.

SINGLE-COMPARTMENT MODELS

Lidwell and Lovelock were apparently among the first to compare concentrations of a pollutant with a mass-balance model. Their model involved the instantaneous introduction of a nonreactive pollutant into a room and considered the dilution resulting from a constant ventilation rate in which the input air was pollutant-free. They noted that, when the air in the room was not well mixed, the dilution by ventilation air was not necessarily exponential; nor were the rates of dilution the same in all parts of the room. A portion of the inlet air stream often tends to bypass part of the room. For instance, when both the inlet and exhaust ducts are on the ceiling, the lower half of the room (and especially the corners) is apparently bypassed and the air in it is diluted more slowly than expected.

Brief⁸ suggested the use of a mixing factor (a constant, usually ranging in value between 1/3 and 1/10, that multiplies the ventilation rate) to account for dilution rates that are lower than would exist if the room air were continually well mixed. Constance¹⁵ also recommended the use of mixing factors. Drivas et al.²² derived mixing factors ranging in value between 0.3 and 0.7, except when fans were used; with fans, the characteristic time for mixing the air throughout the room was short, compared with the characteristic residence time, and the mixing factors were close to unity.

 $m Milly^{65}_{}$ used a single-compartment model involving the instantaneous introduction of a nonreactive contaminant with a pollutant-free input air stream in his discussion of chemical attack of tanks and fortifications. Calder $m ^{11}_{}$ used a single-compartment model in his analysis of the protection afforded by buildings against biologic-warfare aerosol attack; he permitted the outside concentration to vary with time and took into account the surface removal of aerosol by means of a first-order sink term; his results can also be used to describe doses associated with radioactive or chemical contaminants. Calder $m ^{12}_{}$ also used a single-compartment model to calculate dosages associated with the penetration of a forest canopy by aerosols. Milly and Thayer $m ^{66}_{}$ developed a technique for predicting indoor dosages of pollutants generated outdoors, on the basis of a single-compartment model.

 $\overline{\text{Turk}_{--}^{101}}$ presented a detailed analysis of the transient behavior of a single-compartment model involving a constant generation term, and a constant outside concentration; he then considered several special cases during his analysis of the measurement of odorous vapors in test chambers.

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Hunt⁴⁴ used a single-compartment model with a constant internal source and a first-order sink term to interpret data regarding airborne dust in post-office facilities. Hunt et al.⁴⁶ and Cote and Holcombe¹⁷ used a single-compartment model in their investigations of nonreactive gaseous pollutants indoors. Bridge and Corn⁷ used a single-compartment model to predict concentrations of carbon monoxide and particles associated with smoking of cigarettes and cigars; their results were in good agreement with measured values. Sabersky et al.⁹⁰ reported that a single-compartment model involving an outdoor concentration that varied sinusoidally in time and a first-order heterogeneous (surface) decomposition term gave qualitative agreement with data for indoor concentrations of ozone. Shair and Heitner⁹⁵ started with a single-compartment model to develop a "linear-dynamic" model by which the indoor concentrations of ozone can be related to those outside by means of a simple expression. Tests conducted with 24 forced-ventilation systems in 13 laboratory-office buildings yielded values of k (the heterogeneous-loss constant) of 0.02–0.08 cm³/cm²-s, with an average of 0.04 cm³/cm²-s.⁹⁴

To save energy, the makeup-air flow rate in buildings has been reduced. In one case, the reduction in the makeup-air flow rate was sufficient to permit economical selective filtering of the makeup airstream (with activated charcoal) during the times when the outdoor air quality was relatively poor (see Figure VI-3); use of the activated-charcoal filters only when needed and replacement of inexpensive prefilters every couple of months extended the life of the activated-charcoal filters to about 3 yr. This system was designed with the aid of the "linear-dynamic" model. Husuda single-compartment model to examine the feasibility of intermittent operation of mechanical ventilation systems with an eye to conserving energy while maintaining acceptable indoor air quality.

Moschandreas (personal communication) used a single-compartment model of air pollution in nonworkplace indoor environments in Baltimore, Washington, D.C., Pittsburgh, Chicago, and Denver. He monitored carbon monoxide, nitric oxide, nitrogen dioxide, sulfur dioxide, ozone, methane, total hydrocarbons, and carbon dioxide continuously for periods of approximately 14 d in each of five detached dwellings (townhouses), six apartment units, two mobile homes, and one school. In addition, there was a 5-d period of monitoring in one hospital. The model discussed by Moschandreas et al. ⁷² ⁷³ explicitly included a chemical-decay term that was validated with the data base just mentioned. Numerical predictions of hourly carbon monoxide, nitric oxide, nitrogen dioxide, carbon dioxide, and nonmethanehydrocarbon concentrations were found to be within 20% of the observed values 80% of the time. The model did less well in predicting the indoor sulfur dioxide and ozone concentrations; this was attributed to the chemical reactivity of the pollutants. A study was implemented to rank the sensitivity in magnitude changes in output of the model caused by perturbation of an input parameter. The ranking of input parameters, in ascending order of sensitivity, is as follows: initial condition and volume of the structure, indoor source, and pollutant decay and air-infiltration rate of the structure. ⁷²

Shair <u>et al.</u> ⁹⁶ used a single-compartment model to describe the moisture content of bathroom air during and after the use of a shower; they found good agreement with experimental results by considering the solid surfaces in the bathroom to be a sink while the shower is on and a source shortly after the shower is turned off.

Repace and Lowrey⁸⁹ have developed a one-compartment model that describes growth equilibrium and decay of tobacco-smoke aerosol under different room mixing conditions.

Hollowell <u>et al. 42</u> have discussed the impact of radon on indoor air quality. Kusuda <u>et al. 53</u> used the available indoor-radon data to develop a single-compartment model with a first-order radioactive-decay term and a constant generation rate:

$$\frac{V^{dC}}{dt} = qC_0 - \lambda VC + VS,$$

(10)

in which V is volume, C_0 is outdoor concentration, and C is indoor concentration. In addition to terms previously defined, there is the radon-decay constant $\lambda=1.258\times10^{-4}/\text{min}$ and the average source strength per unit volume of air, S. Setting the left-hand side of the above equation to zero and solving for the air-exchange rate yields:

$$q/V = (S - \lambda C)/(C - C_0)$$
. (11)

To facilitate determination of effective radon source strengths, future measurements of indoor-radon concentrations should be accompanied by corresponding outdoor measurements and air-exchange rates, as determined, for example, by a tracer-dilution technique.

SUMMARY AND CONCLUSIONS

The main purpose of an indoor air quality model is to show the relationships of indoor pollutant concentrations to those outside, to geometric and ventilation characteristics of a structure, and to internal sources and sinks.

When the characteristic time for mixing throughout the region of interest is short, compared with the characteristic residence time, the region can be considered as a well-mixed "compartment." Even when that criterion is not met, the uncertainty in the values of the ventilation, source, and sink parameters (with the difficulty of doing otherwise) usually does not justify the development of a more sophisticated model.

Consequently, the starting point for essentially all indoor air pollution models has so far been a first-order differential equation representing a pollutant mass balance in a compartment. In many cases, only the steady-state solution is needed, and the model reduces to an algebraic equation. In a few cases, two or more compartments (usually coupled by internal ventilation streams) have been used to develop a model of indoor air pollution. The main difference between various models arises from choosing different source or sink mechanisms.

To obtain as much general information as possible, the researcher should include the following in the model: corresponding outside concentrations, appropriate geometric parameters, reasonably accurate ventilation rates, and, if possible, sink and source strengths. Such information is required for any model based on a pollutant mass balance.

ESTIMATION OF TOTAL EXPOSURE TO AIR POLLUTION

Today's data on urban air quality come mostly from measurements at fixed monitoring stations. Such data probably show accurately the exposure of a hypothetical person who spends all his time at the station's intake probe. However, people are in constant motion in urban areas, moving from residential areas to places of work to commercial areas, etc. To determine individual human exposure to air pollution accurately, it is necessary to find some means to measure and correlate the movement of individuals in a population and the spatial variation in concentrations of pollutants, whether indoors or outdoors.

One way to estimate better the total individual exposure to environmental pollutants is to equip a large number of persons with monitoring instruments and allow them to go about their daily

activities in a normal manner. However, no large-scale personal monitoring studies have been done or are in progress, at least partly because the development of total-exposure monitoring is still in an early stage. Although no large-scale national program to develop personal monitors has evolved, limited funds from federal agencies have resulted in the development of specific monitors, and private companies have developed some instruments that are portable, small, and reasonably priced. $^{19\ 69\ 76}$

Other approaches have used theoretical analyses and models for estimating total exposure. Fugas 27 made one of the first attempts to compute total exposure from experimental data; her approach was intended only as an illustrative example. She obtained measurements of average concentrations of lead, manganese, and sulfur dioxide during the winter of 1972–1973 from official air-monitoring stations in the city. The measurements were taken at the breathing zone in several streets during business hours, indoors close to the streets during business hours, and in the countryside. By estimating the time spent by inhabitants of the city in five locations—home, work, street 1, street 2, and the countryside—Fugaš calculated the "weighted weekly exposure" (WWE) for each of these air pollutants (see Table VI-2). An intermediate computation is the "integrated exposure," which is the product of the average concentration and the time during which pollution occurs. To calculate the WWE for sulfur dioxide, for example, we note that a person spent an average of 110 h/wk at home, where the average concentration was 89 μ g/m³, for an integrated exposure of 9,790 μ g-h/m³ for the time spent at home. By adding all the integrated exposure components, Fugaš obtained the total of 16,896 μ g-h/m³ for the week. The WWE to sulfur dioxide was then obtained by dividing by the number of hours in a week: 16,896/168=101 μ g/m³.

Duan²³ has modified Fugaš's approach by substituting the term "microenvironment types" for the "locations" used to compute WWE. In Duan's model, a person's integrated exposure over some period (for example, a week) is computed as a weighted average of the exposures from various microenvironment types, weighted by the proportion of time spent in each microenvironment type:

$$E_{ij} = \sum_{k=1}^{K} C_{ijk} t_{ijk},$$

where E_{ij} is the integrated exposure of the ith individual during the jth period, C_{ijk} is the average concentration in the kth microenvironment type during the jth period, and t_{ijk} is the activity-pattern coefficient denoting the time the ith individual spent in the kth microenvironment type during the jth period. Duan²³ suggested that a microenvironment type should be defined "finely" enough to be homogeneous; that is, the concentration coefficients should not vary appreciably over the individuals. However, the microenvironment types have to be somewhat "coarse," so that the analyst will not have too many types to deal with. Some types might be "rush-hour highway commuting," "daytime urban office with air-conditioning," and "weekend daytime outdoors in the park."

Moschandreas and Morse 70 have suggested an analogous approach for computing air-pollution exposure and have applied it to real data. They introduced the application of "mobility patterns," which are designed to capture "the daily movements of individuals as they move to and from work, from home to points of amusements, adventure, business, and so on." By examining the literature on activity patterns and "time budgets," they arrived at the estimated time that all persons—all races, ages, socioeconomic groups, workers, students, etc.—spend in various "environmental modes" (which are analogous to the "microenvironment types" of Duan 23 and the "locations" of Fugas 27). The

population spends 72.8% of its time inside homes, but the figure is different for different population subgroups (workers, children, the elderly, etc.).

Because of the importance of considering the mobility patterns of population subgroups, Moschandreas and Morse²¹ examined U.S. census data to define the percentage of each of six subgroups in the total population: housewives, office personnel, industrial workers, outdoor workers, elderly and infirm people, and students. However, students are not considered in the overall model, because few studies have been made of their mobility patterns. The model can be viewed as a threedimensional drawing in which persons move through time occupying different environmental modes, thereby exposing themselves to the particular concentration that is associated with each environmental mode and period. On the basis of data on typical diurnal ozone concentrations for three environmental modes (residential, office, and indoors) in the Boston area and estimates of the percentage of the population in each environmental mode as a function of time, Moschandreas and Morse⁷⁰ estimated that 21% of the population is exposed to ozone at 80 ppb or more. At the time, the federal NAAQS for ozone (1-h average) was 80 ppb, but it has since been raised. Among individual population subgroups, Moschandreas and Morse estimated that outdoor workers are exposed to high ozone concentrations (over 80 ppb) for about 6 h and industrial workers for the 1-h period between 4:00 and 5:00 p.m. If we take a "snapshot" of the estimated population exposures at some particular point in time, such as 3:00 p.m. (that is, 1500; see Figure VI-4), some 25% of the population (3.9% in outdoor activities and 21.3% in transit) is exposed to ozone at 80 ppb or more.

Although these are excellent recent examples of ways to estimate population exposure to air pollution, they have several limitations. Health-related air quality standards usually do not have weekly averaging times. Thus, the value of the WWE calculated by Fugaš²⁷ cannot be compared directly with existing air quality standards. Duan's formulation²³ was intended to be more flexible, allowing any averaging period (for example, a week or a month) to be used. The computation of exposure by Moschandreas and Morse⁷⁰ did not cause difficulties with averaging periods, because ozone has a 1-h NAAQS, and the population is assumed to spend 1-h increments (or multiples of 1 h), in each environmental mode.

Each of the above approaches for calculating exposure gives the population's <u>average exposure</u> over some specified period, and a problem arises from the emphasis on the arithmetic mean. In any given period, some people will be involved in combinations of activities that can result in exposures much higher and much lower than the mean. In addition, the time spent in each activity varies from day to day and from person to person. Thus, the mean value for exposure concentration is not adequate to characterize the highest concentrations to which members of the population are exposed, and the variance of exposures also must be considered. Ideally, there would be an effective technique for determining the entire frequency distribution of exposures of the population to air pollution.

As discussed above, two approaches are used for estimating the frequency distribution of human airpollution exposures: modeling, which relates the activities of persons as a function of time and the concentrations to which they are exposed; and field studies, which use personal monitors to cover a large enough population sample (or a stratified sample) to represent statistically the distribution of exposures. Although a large-scale field study of exposure has not been completed, efforts are under way to use computer simulation to model human exposures to air pollution.

Ott 77 78 has developed a computer-simulation model of human exposure to air pollution that includes the movements of individual people in a metropolitan area as a series of transitions from one "microenvironment" to another. Ott's computer program, "Simulation of Human Air Pollution Exposures" (SHAPE), uses probability distributions of the time that people spend in each microenvironment—distributions derived from studies of human activity patterns. In each microenvironment, the concentration to which an individual is exposed is treated stochastically, with distributional models that are based on field studies of air-pollutant concentrations reported in the

research literature. In the simulation, the computer keeps track of the exposure received by each person as he or she moves forward in time and occupies successive microenvironments.

 Ott^{77} defines an "exposure" of person i to concentration c statistically as the joint occurrence of two independent events: person i is present in microenvironment j, and the concentration C(j) = c occurs in microenvironment j. C(j) denotes the probability distribution of the concentrations associated with the microenvironment and is based on field monitoring data. Then, the integrated exposure ϵ_i of person i is computed as the sum of the products of the concentrations encountered in each microenvironment and the time spent there:

$$\underline{\varepsilon}_{i} = \sum_{j=1}^{J} c_{j} t_{ij},$$

where c_j is the concentration associated with microenvironment j, t_{ij} is the time spent by person i in microenvironment j, and J is the total number of microenvironments occupied by person i in some period of interest. Conceptually, this model can be represented by a three-dimensional array (see Figure VI-5) that is similar to a three-dimensional space developed by Moschandreas and Morse. However, the computer simulation follows one person at a time through his or her daily activities, and the resulting distribution of exposures is obtained by considering those of all persons in the simulation. The times t_{ij} spent in the microenvironments are variable and do not need to be integer multiples of 1 h.

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Figures

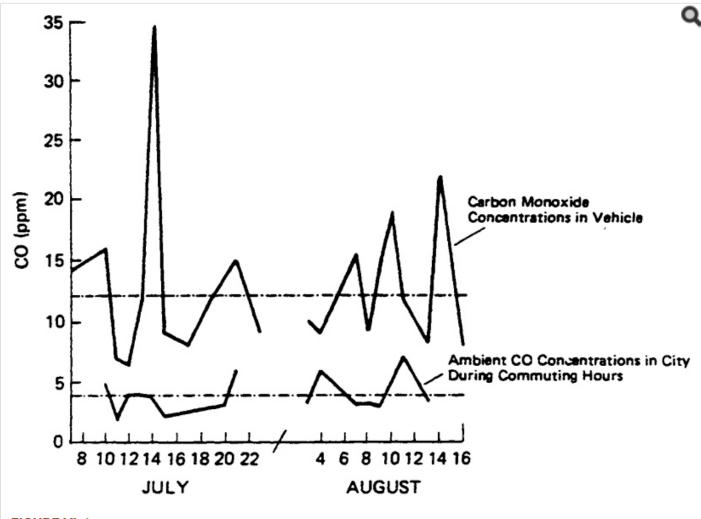
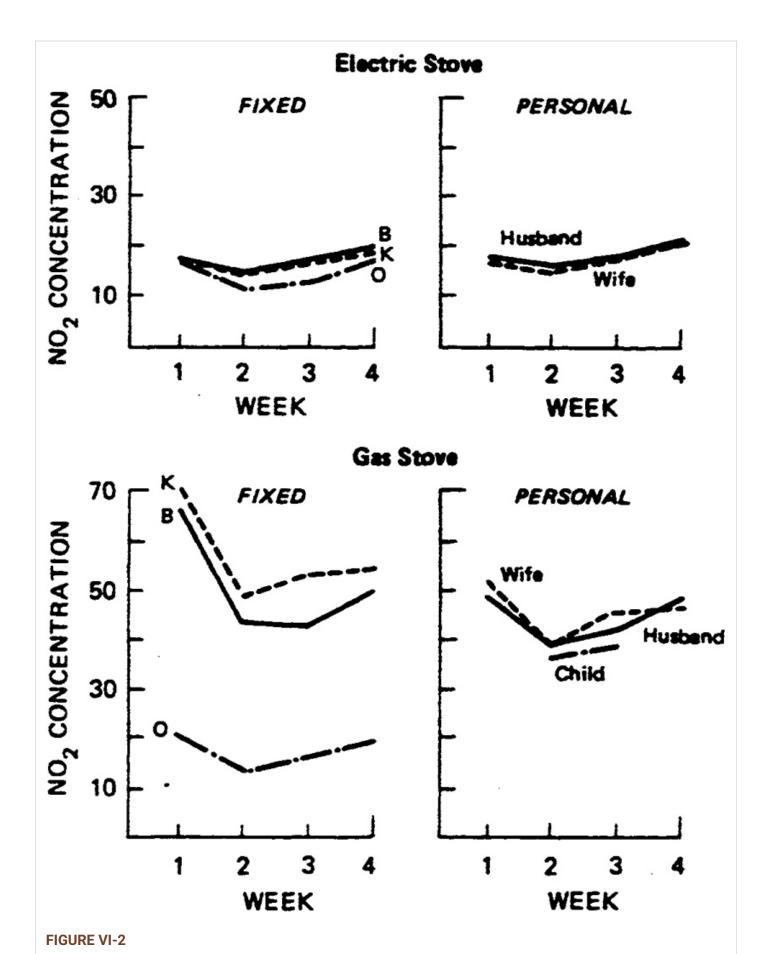


FIGURE VI-1

Carbon monoxide concentrations in vehicle, compared with ambient concentrations in city. Reprinted from Wallace. 104



Week-long nitrogen dioxide concentrations, $\mu g/m^3$, for an electric-cooking and a gas-cooking family in Topeka, Kansas, May-June 1979. B, bedroom; K, kitchen: O, outdoor. Reprinted with

permission from Dockery <u>et al</u>.

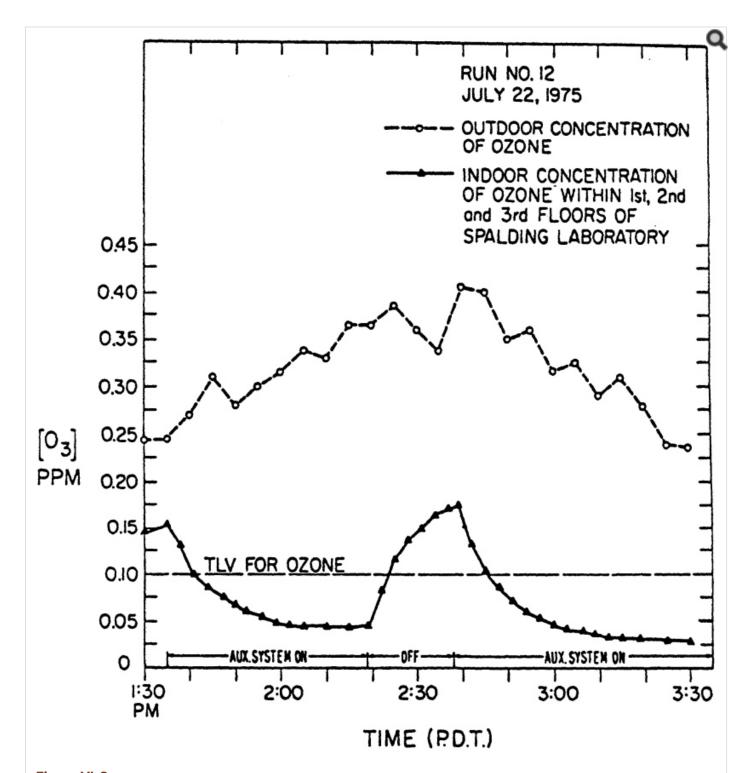
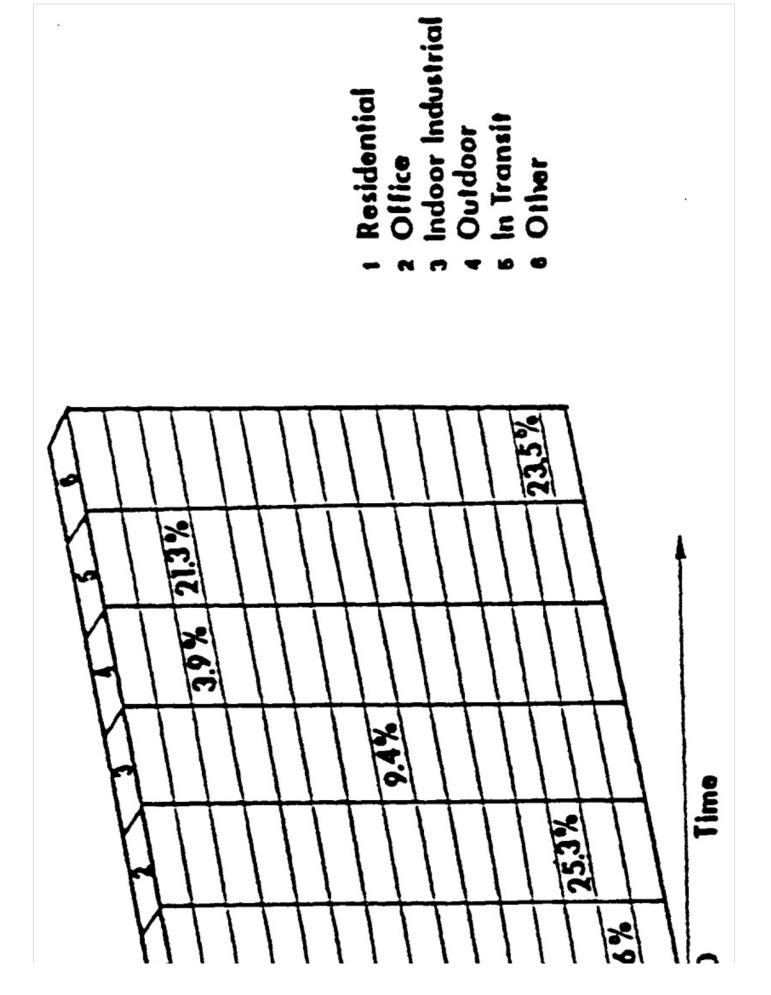


Figure VI-3
Relative indoor and outdoor ozone concentrations. From F.H.Shair (personal communication).



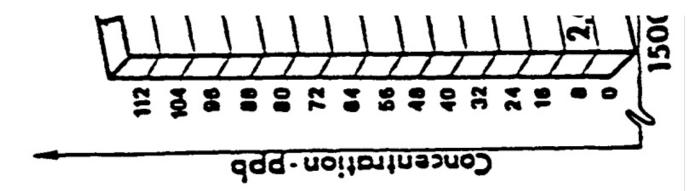


FIGURE VI-4

Example showing proportion of population exposed to hourly ozone concentrations during the 1500 hour (3:00 p.m.) in each environmental mode (numbered above). Reprinted with permission from Moschandreas and Morse. $\stackrel{70}{_}$

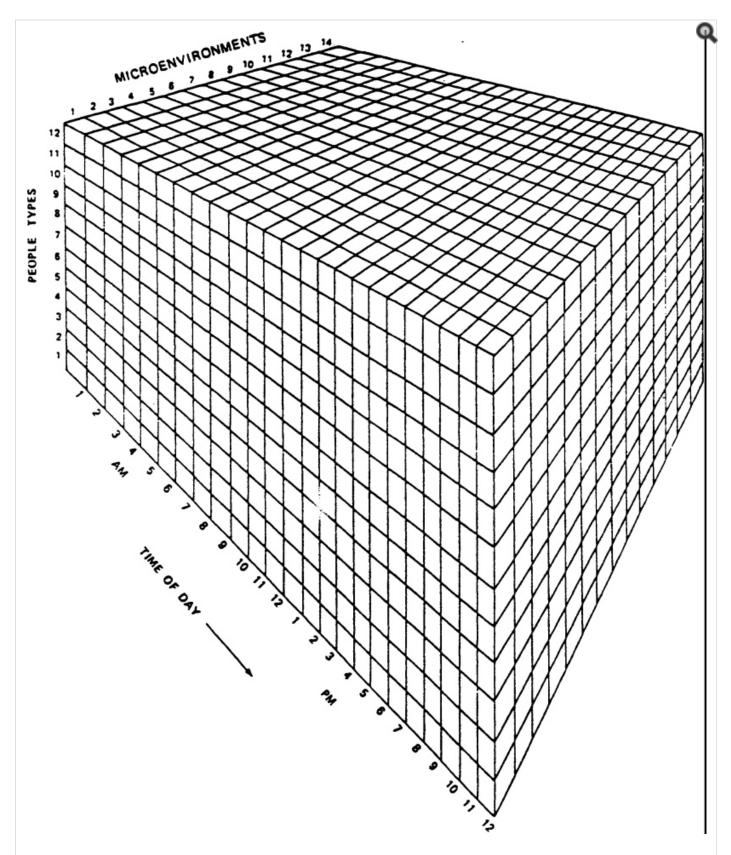


FIGURE VI-5

Graphic representation of person-environment-time array in computer simulation model of exposure to air pollution suggested by Ott. $^{77}_{-}$

Tables

TABLE VI-1 Calculation of Time-Weighted Weekly Average Exposure of Office Worker in Zagreb to Airborne Lead $^{\rm a}$

Location or Activity	Average Lead Concentration, μg/m ³	Duration of Exposure, h/wk	Integrated Exposure, µg-h/m ³		
Workplace	1.2	42			
Outdoor activities	6.3	14	88.2		
Recreation	0.2	6	1.2		
At home:					
Rest of day	0.7	22	15.4		
Night	0.3	48	14.4		
Weekends	0.5	36	18.0		
Total	_	168	187.6		
Weighted-averag	ge weekly exposure: 1.1 μg/m ³				

a Data from Fugaš <u>et al</u>.__

TABLE VI-2 Example by Fugaš Illustrating Computation of Weighted Weekly Exposure a

Type of Exposure	Duration of Exposure, h/wk	Sulfur Dioxide		Lead		Manganese	
		С	Ct	c	Ct	С	Ct
Home	110	89	9,790	2.5	275	0.04	4.4
Work	42	8	336	0.3	12.6	0.02	0.84
Street 1	10	600	6,000	6.0	60	0.80	8.0
Street 2	4	180	720	3.5	14	0.12	0.48
Countryside	2	25	50	0.1	0.2	0.01	0.02
Total	168	_	16,896	_	361.8	_	13.74
Weighted weekly exposure	_	_	101	_	2.2		0.08

a Data from Fugaš. 27 Values for C (concentration) are expressed in $\mu g/m^3$, and values for Ct (integrated exposure) are expressed in $\mu g-h/m^3$.

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