
Techniques to Estimate Potential Exposures to Chemicals in the Environment

JOHN D. SPENGLER and LANCE A. WALLACE

1 INTRODUCTION

In the last two decades, there has been a tremendous advance in analytical devices. With these field and laboratory instruments, our understanding of the composition and behaviour of the natural environment has rapidly advanced. Size fractionated particles can now be routinely analysed for their elemental, ionic, and carbonaceous composition. These techniques are adequately described in several publications, among them: *Analysis of Airborne Particles by Physical Methods*, Malissa, Ed. CRC Press, 1970; and *Particulate Carbon: Atmospheric Life Cycle*, Wolff and Klimisch, Eds. Plenum, 1982. From measurements using these techniques, we now have evidence of long-range transport of aerosols even into the remote regions of the world. Using statistical inference methods the receptor-based information contained in a set of samples can yield descriptions of source contributions.

While spectroscopy, electrochemical, chemiluminescent, and gas chromatographic principles have been applied to a variety of gases, most commercially available instruments have been oriented to concentrations experienced in urban areas or stacks. The emphasis has been towards the criteria pollutants of O_3 , NO_2 , SO_2 , and CO . However, recently more attention has been given to an array of organic materials: formaldehyde from resins; alkanes and aldehydes from woodburning and vehicle exhaust; halogenated hydrocarbons from solvents, adhesives, and consumer products; and polynuclear aromatics and nitrosamines from combustion.

It is clear that, with the assistance of laboratory-based analytical instruments, accurate and precise measurements can be made of most environmental contaminants of current interest at very low levels. However, if the purpose of environmental assessment is to provide reliable measures of human exposures, then the question of available techniques and instruments has to be viewed in this particular context. Because of the mobility of the population, throughout the day and over a lifetime, we

experience different concentrations and mixtures of contaminants in many microenvironments.

The time/use patterns of humans vary by age, sex, season, socio-economic class, ethnic background, climate, weather, among many other factors. The concentrations in these microenvironments may also vary in time, because emission, dilution, transport, chemical reactions, and removal processes are dynamic. Even within similar classes of microenvironments, concentrations can be substantially different. Because of these factors, the concentrations measured by fixed monitors or even by a finite integrated sample may not represent the concentration in the immediate boundary of a person. It is these proximate concentrations that constitute the actual human exposure. Exposure commonly refers to concentrations of contaminants measured in a specific environment. However, unless these concentrations refer to measurements in air, water, food, soils, liquids that are inhaled, ingested, or absorbed through the skin (Ott, 1990), then they are just that—concentrations as surrogates for actual human exposure.

On passing the boundary of the body, these exposures become the dose. Two distinctions can be made about dose: The “internal” dose is the amount of a substance or its metabolites in tissue; and the “biologically effective” is the amount that interacts with a particular target tissue. Hence, dose involves a variety of host factors that encompass the concept of uptake. It is important to understand these distinctions. Site- and time-specific concentrations, while perhaps relevant to soiling, deposition, or ecological damage, may be irrelevant or limited with respect to human exposures. Further, depending on the specific purpose, assessment of human exposures should consider the time, concentration, and host-dependent factors relevant to the biochemical mechanisms implicit in the term “dose.” Some contaminants may manifest worse effects from short-term peaks than from longer exposures with the same integrated value. On the other hand, some contaminants have cumulative effects while still others have age-dependent or synergistic factors influencing the health consequences of environmental exposures.

This paper will deal almost exclusively with instruments and techniques relevant to air pollutant exposures, notwithstanding the previous definitions. When researchers discuss air pollution exposures they are referring to concentration measurements either in the microenvironments frequented by people or to measurements made with personal monitoring equipment carried or worn.

2 INSTRUMENTATION FOR PERSONAL EXPOSURES

Before reviewing available instrumentation for personal monitoring, it is appropriate to discuss the purposes of exposure assessment. Depending on the purpose, the design of the study and the characteristics of portability, response time, sensitivity, precision, and accuracy required of the instrumen-

tation will vary. While some studies have multiple objectives, the selection of subjects, the duration of monitoring, and the integration time of measurement may vary. As will be discussed, currently available instruments will not provide the opportunities to address human exposures to most air pollutants over time-scales of interest.

Essentially, the purpose of personal exposure studies can be grouped around one of three major objectives:

- (1) Exploratory investigations to broaden our understanding of the actual dynamic range of exposures (this may be a justifiable objective by itself or done in conjunction with a pre-assessment to design epidemiological studies);

- (2) Utilisation for health risk analysis and epidemiology to identify target populations or to quantify the nature of the exposure/response functions (here, the issues of misclassification of exposure are relevant to sampling size, identifying confounding influences, and to reducing uncertainty in analysis results);

- (3) Targeting control strategies for source reduction or receptor modification to quantify the net change in exposures to the population or to a specific subgroup.

The study of personal exposures to air pollutants is important to epidemiological studies, standard setting, and regulation. Advances have been made in our understanding of personal exposures to a few pollutants, specifically, carbon monoxide, respirable particulates, volatile organic substances, nitrogen dioxide, and lead (Spengler and Soczek, 1984).

To obtain data on personal exposures, either a direct or an indirect scheme may be used. Direct assessment invokes the measurement of an individual's exposure with a personal monitor. These monitors are almost always integrating samplers, although continuous analysers for carbon monoxide and respirable particles have been recently introduced that can be used as personal monitors. Rapid response analysers for most pollutants, sensitive in the ranges of concentrations typically encountered in non-industrial settings, are available but not sufficiently portable.

If individuals cannot be monitored, an indirect approach may be implemented. With knowledge of the time/activity patterns of individuals or populations and knowledge of the concentrations in the microenvironments or locations that they frequent, one can calculate their integrated exposures.

While this approach does not involve personal monitors that individuals actually carry, it still requires lightweight portable instrumentation to obtain the concentration profiles in the locations frequented by the individuals.

To date, information on the distributions of personal exposures is limited. These limitations are as follows:

- (1) Only a few pollutants have been studied. For the most part, the list has been limited to those pollutants for which monitors are available: CO, NO₂, particles, Pb, volatile organics and semivolatile organics (pesticides and polyaromatic hydrocarbons).

- (2) Time resolutions of most personal exposure studies are typically of

the order of days or perhaps hours. There is little information available on the short-term peak or the long-term exposures to which individuals are exposed.

(3) Only a small segment of society has actually been studied. Those study populations can be characterised for the most part of white, urban or suburban, middle-class adults. Those that are employed usually perform non-manual (white-collar) labour. Few children or infants have been studied.

(4) The locations that have been monitored have primarily been homes and some office buildings. Even within these structures we do not have good resolution of concentrations as they vary temporally and spatially. There is little information available on actual in-vehicle exposures during transportation or on occupationally related exposures.

(5) There have been relatively few advances in personal monitoring instruments. Most instruments are passive monitors that integrate exposures over hours to days. Since there is no regulatory requirement to stimulate commercial development, personal monitoring equipment suitable for personal exposure studies on the general public is not readily available. The devices available (with few exceptions) are developed by researchers for specific studies. This, in turn, can lead to problems comparing studies reporting exposures to the same pollutant but using instruments that have not been evaluated or compared with one another.

3 SAMPLING EQUIPMENT

A personal monitor is defined as an instrument that can be carried or worn by a member of the general public. Thus, it should be light (<1 kg), quiet, self-powered for a minimum of 12 hours, and non-bulky. A portable monitor suitable for micro-environmental measurements can be much heavier (<20 kg), somewhat noisier, may require a line source of power, and may be quite bulky.

The gases of interest are carbon monoxide, nitrogen dioxide, volatile organic vapours such as benzene, nitrosamines, and aromatic and halogenated compounds and semivolatile organic compounds such as pesticides. These gases may have indoor and outdoor sources. Other gases, such as ozone, sulfur dioxide, and hydrogen sulfide, are primarily limited to outdoor or occupational environments. While penetration into indoor environments will modify pollutant concentrations, assessment of personal exposures to these last-named gases might be better gauged by an indirect method in which the indoor to outdoor relationships are developed.

Suspended particles in the atmosphere can be characterised by size, shape, mass concentration, number concentration, chemical composition, elemental composition, solubility, and acidity among other attributes. It is useful to distinguish particles by size and composition. Under typical situations, they tend to be bimodally distributed in the atmosphere by mass/size

concentrations. This is a useful distinction, because separating particle by size according to coarse fraction ($d > 2.5 \mu\text{m}$) and fine fraction ($d < 2.5 \mu\text{m}$) provides the basis for distinguishing between many sources. Combustion and gas-to-aerosol conversion generate particles in the fine fraction. Actually, most of the secondary aerosols including acidic particles, condensed metal and organic vapours are associated with particles less than $1 \mu\text{m}$. Grinding and abrasive processes produce larger particles.

Fugitive industrial emissions and eroded soils and surfaces generate particles typically greater than $2 \mu\text{m}$ in diameter. The human respiratory tract separates particles into similar sizes. The fine fraction particles have a greater likelihood of penetrating to the distant airways and alveoli. Therefore, the principle of sampling particles, as well as the properties measured, are important to concepts of exposure and dose.

Most personal sampling equipment uses low flow filtration, and measures the mass of particles collected. Various filter substrates can be used depending on the physical parameters of the sampler (size, flow rate, pressure drop). Filter medium is an important consideration, if subsequent chemical, elemental, morphological, or acidity analyses are planned for the collected particles.

Beyond the issues of filter blank and uniformity of deposition are the concerns for stability and preservation of the sample collected. Reactive particles can react on the filter with other species, with gases in the atmosphere, or can volatilise after collection. This subject is treated in more detail elsewhere (Stevens, 1984).

Other properties of particles can provide measures of the mass and composition. Only light scattering by submicron size particles has been exploited to the point of commercialisation of light weight portable/personal equipment. These instruments will be discussed in this chapter.

3.1 CARBON MONOXIDE (CO)

Among inorganic gases, carbon monoxide has been most often measured in personal exposure studies. Several instruments capable of measuring CO at environmental levels are available (Gruber *et al.*, 1979; Forlenza, 1983; Stetter *et al.* 1979; Kosek and LaConti, 1983). They may be classed as active (pump required) or passive (diffusion) devices. In the United States, direct-reading electrochemical devices of both types are available at prices ranging from about \$1,000 to \$2,000. However, in no case has adequate testing of these devices at environmental levels occurred. One large study of 1200 persons in Denver (Johnson, 1983) and Washington D.C. (Hartwell *et al.*, 1984) has been carried out using the GE CO detector and a data logger designed by EPA scientists (Ott *et al.*, 1984).

Carbon monoxide has often been used as a surrogate measure of cigarette smoke (Hoffmann *et al.*, 1979a) primarily because of the ready availability of portable instruments such as the Ecolyzer. The levels of CO are not, however, raised sufficiently above background levels to serve as an unequivocal measure of smoking activity. For example, the EPA study mentioned above

showed that passive smoking raised CO levels by an average of 1.5 ppm above a background level of approximately 3 ppm. The same study showed that use of the gas stove raised CO levels by 2.5 ppm; thus, cooking with gas is a powerful confounder, if smoking exposure is measured using CO as a surrogate. Finally, the study showed that temperature, humidity, and especially zero drift effects can cause large errors at usual environmental levels of 0 to 3 ppm (Wallace *et al.*, 1984c). For these reasons, measurement of CO as a surrogate for smoking exposure is not recommended except in cases where no other measurement techniques are available.

3.2 NITROGEN DIOXIDE

Important sources of NO₂ are automobile exhaust and gas stoves. NO₂ has occasionally been measured by personal monitors in studies involving passive smoking (Melia *et al.*, 1977, 1982; Goldstein *et al.*, 1979; Comstock *et al.*, 1981; Good *et al.*, 1982; Matsushita and Mori, 1984; Nitta and Maeda, 1982). The most popular personal NO₂ monitors are the Palmes tube (Palmes *et al.*, 1976) and Yanagisawa badge (Yanagisawa and Nishimura, 1982). Both devices rely on diffusion and a triethanolamine (TEA) absorbent to collect NO₂, followed by spectrophotometric analysis. The Palmes tube employs metal grids soaked in TEA, while the badge uses a membrane impregnated with TEA. The badge has a 5-layer baffle to reduce velocity effects and is about 10 times more sensitive (66 ppb-h) than the tube. The tube has been employed in 3-day, 7-day, and 2-week integration modes (Spengler *et al.*, 1983), while the badge could be used for periods as short as one day. Direct-reading continuous monitors are available for levels about 10 ppb and greater. However, these electrochemical and chemiluminescent devices have been shown to have interferences when used indoors. Nitrous acid vapours among other gases limits the use of these devices as an absolute measure of NO₂.

3.3 VOLATILE ORGANICS

Personal monitors employing Tenax have been used in EPA's TEAM Study to measure exposures and corresponding breath levels of about 700 persons in 10 U.S. cities (Pellizzari *et al.*, 1984c; Wallace *et al.*, 1985a,b, 1987, 1988). The monitors consist of a pump capable of operating for about 12 hours at about 30 ml/min flow rate and a glass tube containing about 2.2 g of 40/60 mesh Tenax. Analysis is by thermal desorption and GC/MS (Pellizzari *et al.*, 1984a, b). Extreme care must be taken to clean the Tenax before use, protect it from contamination and/or loss during transport and storage, analyse samples promptly, and include extensive quality control procedures such as spiking samples with deuterated compounds to detect losses. The monitors have been used in studies of 350 residents of New Jersey and 200 residents of California. Identical samplers were used in an EPA study of organics in 10 commercial buildings (Sheldon *et al.*, 1989a,b). Tenax and other sorbents were also used

in a study of 15 homes in Northern Italy (de Bortoli *et al.*, 1984).

Indoor sampling of 135 homes in the Netherlands (Lebret, 1986) and 500 homes in Germany (Krause, 1987) has been carried out using activated charcoal. Results agreed closely with the results using Tenax—namely, that indoor concentrations of about 20 to 30 organic vapours were consistently elevated, in some cases by an order of magnitude, over outdoor concentrations. This finding has great importance for future epidemiological studies of these compounds, virtually requiring some form of personal or, at least, indoor monitoring to gain an adequate measure of exposure.

3.4 FORMALDEHYDE

Monitoring of formaldehyde is extremely difficult because of the influence of temperature, humidity, and history upon its emission. Also, due to the high level of public interest, it warrants individual attention.

New passive monitors give promise of being useful. One, developed at Oak Ridge National Laboratory, employs a permeable dimethylsilicone membrane. The collection medium is water or a 13 "X" molecular sieve. Both collect formaldehyde vapour efficiently, but the sieve is more stable during longer sampling periods and longer delays between sampling and analysis. Analysis is by either the MBTH (3-methyl-2-benzothiazolinone) or *para*-rosaniline methods. A detection limit of about 150 ppb-h and a quantifiable limit of about 500 ppb-h are attainable for the 7.9 cm² sampler using the MBTH method.

A second monitor, developed by Lawrence Berkeley National Laboratory, uses a glass fibre filter treated with sodium bisulfite contained in a glass vial. The exposed filter is removed, distilled water is added to the filter, and the resulting solution is analysed by the chromotropic acid/spectrophotometric procedure recommended by NIOSH. This device is less sensitive than the Oak Ridge monitor, with a detection limit of about 10 to 20 ppm-h.

The 3M company has developed a diffusion badge capable of measuring quantitatively formaldehyde exposures as low as 770 ppb-h (i.e., 32 ppb over a 24-hour monitoring period). The badge clips on the wearer's lapel for personal sampling and can be attached to a surface (away from walls or ceilings) for sampling indoors. Formaldehyde vapours are absorbed on bisulfite-impregnated paper.

The monitor is desorbed using formaldehyde-free distilled water. Aliquots of the desorption solution are reacted with freshly made chromotropic acid in the presence of sulfuric acid. A purple monocationic chromogen is formed. The absorbance of the coloured solution is read in a spectrophotometer at 580 nanometers.

3.5 PESTICIDES AND PCBs

Personal exposure to airborne organochlorine pesticides and PCBs has been measured using a personal monitor employing polyurethane foam (PUF).

A medium flow pump pulls air at 2 to 4 l/min across the PUF cartridge for an 8- to 12-hour period. Analysis is by gas chromatography using electron-capture detection (GC/ECD). The method has been validated in laboratory studies for 20 organochlorine and carbamate pesticides. It has been employed in an EPA study of personal exposures to 31 household pesticides and termiticides of 250 residents of Springfield, Massachusetts, and Jackson, Florida (Immerman, 1990). The banned termiticides heptachlar, chlordane, aldrin, and dieldrin were found to present the highest risk, perhaps through track-in of contaminated soil. Thus the exposure of children may require measurement techniques for household dust, soil ingestion, and dermal absorption as well as for indoor air.

3.6 PARTICLES

Personal and portable monitors for measuring particle mass have been used in a number of studies, notably those of Spengler and co-workers (Repace and Lowry, 1980; Dockery and Spengler, 1981; Colome *et al.*, 1982; Quant *et al.*, 1982). Devices employed usually include gravimetric (pump-filter) for long-term sampling, and piezoelectric or optical methods for near real-time sampling.

Gravimetric devices requirements for a personal monitor include a pump capable of collecting about 4 l/min for about 10–12 hours on a single battery charge. (This flow rate is a minimum to allow accurate analysis, based on likely environmental concentrations.) Only a few such monitors exist (Fletcher, 1984) including the Harvard-EPRI cyclone sampler (Turner *et al.*, 1979) and the EPA-NBS stack-filter sampler (Bright and Fletcher, 1983).

The Harvard-EPRI sampler utilizes a 10 mm cyclone to remove large particles ($> 10 \mu\text{m}$), a long-life Brailsford brushless pump, a battery capable of 14 to 20 hours operation, and a self-contained battery charger. Flow rate is presettable at 0.5 to 3 (± 0.1) l/min. A pulsation dampener is required. Filter cassettes containing a 37 mm Fluoropore filter with a $1 \mu\text{m}$ pore size allow simple replacement and automated analysis.

The EPA/NBS sampler was created by the National Bureau of Standards to EPA specifications, which included a requirement to collect both the fine ($< 2.5 \mu\text{m}$) and coarse ($> 2.5 \mu\text{m}$) fractions of inhalable particulate matter. The monitor employs replaceable nozzles capable of providing variable upper diameter cutoffs of 7, 10, and $15 \mu\text{m}$. A Bendix pump powered by Gould Ni/Cd batteries provides 6 l/min flow rates for 24 to 40 hours. A $6.8 \mu\text{m}$ pore size Apiezon-L-grease-coated Nucleopore filter is used to collect the coarse fraction and a $3 \mu\text{m}$ pore size Zefluor filter for the fine fraction. Tests showed that this filter collects 98 percent of the fine fraction, while still presenting minimum (4 to 5 in of water) pressure drop at 6 l/min. 24-hour test runs with EPA dichotomous samplers showed good agreement between both the fine and coarse fractions collected by each instrument.

A portable monitor suitable for automated sampling of the fine and coarse fractions of inhalable particulates in indoor environments for up to two-

week periods has been recently developed at the Harvard School of Public Health (Turner *et al.*, 1984). The monitor includes an impactor inlet to separate the inhalable particulates ($<10\text{ }\mu\text{m}$) from larger particles. The impactor can accept 42 mm or 37 mm filters in a "2 × 2" removable slide holder. The impactor deposits the particles uniformly ($<10\%$ variation) on the filters for X-ray fluorescence analysis. Gravimetric and beta-ray attenuation measurements can also be performed. The monitor has several attractive design features, including a preset timer allowing one to select predetermined periods (4 hour minimum) of sampling over a two-week duration. A quiet Japanese-made pump samples air at 4 l/min off-line power.

A piezoelectric portable monitor, the Piezobalance of Thermosystems, Incorporated (TSI), has been used in investigations of exposure to particles indoors. The principle involves comparing the frequency of a piezoelectric crystal exposed to the atmosphere with that of a matched unexposed reference crystal (Daley and Lundgren, 1975; Sem and Tsurubayashi, 1975; Sem *et al.*, 1977). A pump pulls air at 1 l/min into a chamber where an electrostatic precipitator deposits the particles on the exposed crystal. The region of linearity of response is limited (about 5 mg); for most environmental applications this is not a serious limitation. A manual self-cleaning operation can be carried out in a few minutes. Temperature and humidity changes have a large effect on the Piezobalance, making it unstable in situations of rapidly fluctuating temperatures or humidities (as in an automobile). However, if care is taken to equilibrate the instrument to an indoor or outdoor environment, it can be useful in most situations. A second major problem, production of particles (perhaps by the precipitator acting on large-molecule gases), has been noted recently (Sem and Quant, 1983). This difficulty can be overcome by employing a HEPA filter and establishing a baseline value for each sampling microenvironment. Previous studies that did not employ this technique may have overestimated the concentration levels by 5 to 30 $\mu\text{g}/\text{m}^3$ in all environments studied.

3.7 POLYCYCLIC AROMATIC HYDROCARBONS

This important class of particle-bound organics contains several carcinogens found at low levels in cigarette smoke (Grimmer *et al.*, 1977) and in other products of combustion, notably wood smoke. To collect sufficient material for analysis, it has been necessary to employ high-volume sampling techniques, unsuitable for personal or even micro-environmental sampling. However, a recent passive device developed at Oak Ridge National Laboratory (Vo-Dinh and Winefordern, 1977; Vo-Dinh and Hooyman, 1979; Vo-Dinh *et al.*, 1980, 1981) may be capable of use at environmental levels. The badge contains an adsorbent doped with a heavy element. Analysis methods are synchronous fluorescence and room-temperature phosphorimetry. A question of sensitivity remains, however.

An important study of airborne concentrations of toxic elements and organic species (ATEOS) was conducted in four communities in New Jersey

(Lioy *et al.*, 1983). This study measured inhalable particulate matter and characterised the extractable organic fractions (Harkov *et al.*, 1984). Applying mutagenicity testing, the various extractable fractions of airborne particulate matter were examined for both indoor and outdoor samples (Lioy *et al.*, 1985; Athenholt *et al.*, 1985; Butler *et al.*, 1987). The ATEOS project illustrates how detailed chemical and bioassay analysis of complex mixtures can be analysed using multivariate statistical techniques to understand the contributions of sources and meteorological factors (Morandi *et al.*, 1987).

4 SUMMARY AND CONCLUSIONS

Personal monitors capable of measuring integrated exposures to CO, NO₂, volatile organics, semivolatile organics, and nitrosamines at environmental levels are fully field-tested. However, personal and portable monitors for particles, particle-bound organics, and nicotine, although recently developed and applied, require further evaluation in objective laboratory and field tests. Finally, personal monitors capable of measuring instantaneous exposure to organics, particles, and NO₂ at environmental levels do not exist.

Thus, the tools are largely available for determining long-term chronic exposures to some carcinogens (benzene, nitrosamines, possibly PAHs). However, no extensive programme for monitoring such exposures has been mounted by any research group or governmental agency. By contrast, short-term acute exposures cannot be measured very accurately by existing personal monitors. Since these short-term effects (annoyance, odour, respiratory irritation, so-called "Sick Building Syndrome") may be important (for both health and economic reasons), it would seem useful to develop or improve instruments capable of measuring short-term peak exposure to NO₂, CO, and particles at a minimum.

5 RECOMMENDATIONS

(1) *Development of Test Protocols.* Investigations of indoor air quality and personal exposures have, for the most part, used instrumentation originally developed for industrial hygiene applications. For a number of reasons, this may not be appropriate. For example, the concentrations of indoor contaminants may have ranges and temporal/spatial variation dissimilar from industrial workplaces. Most often, concentrations in homes and offices are lower than those for which the equipment was originally designed. To accommodate this, the equipment may be required to operate at higher flow rates or for longer durations. At these low concentrations, the effects of interfering gases, fluctuating temperatures, and changes in humidity encountered in vehicles, residences, and other types of structures have not been adequately investigated. Therefore, test protocols should be developed for evaluating equipment to be used as indoor/personal monitors. These

devices should be evaluated over the range of anticipated concentrations addressing the possible effects of temperature, humidity, and interfering compounds. This work is urgently needed for integrating and continuous monitors for NO_2 , CO, and formaldehyde. Interfering compounds that should be considered include tobacco smoke, organic and nitrogenated compounds, ammonia and other volatile compounds present in cleaning agents, solvents found in paints and varnishes, and aerosol-spray propellants.

(2) *Portable, Continuous NO_2 Monitors.* Available integrating samplers for NO_2 (Palmer tube, filter badge) are inadequate for fully understanding personal exposures to NO_2 . Sources of NO_2 such as combustion appliances and vehicles are usually intermittent, providing short, intense exposures. The exposures to and subsequent health or irritant effects of elevated peak concentrations of NO_2 are currently unresolved. To measure the former, lightweight personal or portable NO_2 monitoring equipment is needed. The instruments should be capable of responding to concentrations over a range of 5 ppb to 1 ppm with a 90 percent response time of 5 seconds. The instrument should be insensitive to CO, NO, CO_2 , ammonia, and the variety of organic compounds commonly found indoors. The effects of variations in temperatures (0 to 30°C) and humidities (10 to 90 percent) must not compromise accuracy in excess of ± 10 percent. The device should be compatible with an electronic data acquisition system.

(3) *Portable Continuous CO Monitors.* Portable continuous monitoring equipment exists for CO. New personal analysers operating by diffusion (rather than active pumping) and electrochemical sensors/detectors must be evaluated. Concerns for interferences, response characteristics, and low velocities across the sensors need to be evaluated.

(4) *Passive CO Survey Sampler.* An inexpensive passive CO sampler is needed for large-scale surveys of homes, offices, garages, arenas, and other indoor sites. As a design objective, the sampler should have a sensitivity of 25 to 50 ppm-hours and be accurate to within ± 10 percent. This is an important challenge for passive diffusion monitors; however, such sensitivity would permit in-vehicle surveys over typical commute times. A sensitivity of 100 ppm-hour would be adequate for surveying buildings for faulty heating/flue systems over a ten-hour nighttime heating cycle. (An evening heating period is of interest, because it is expected to represent the situation that might maximise exposures). A 10-hour average concentration of 10 ppm would indicate a faulty combustion system. There is a tremendous need for inexpensive passive monitors for measuring indoor exposures in developing countries. CO is a particular concern for countries where charcoal is used extensively for cooking and heating.

(5) *Particulate Sampler.* Respirable particulate matter can be measured with several devices currently available. A 10-mm nylon cyclone pre-separator operating at 1.7 l/min is commonly used in industrial environments to provide the size-fractionation prior to filtration. Sampling times of 12 to 24 hours or more are needed to obtain sufficient mass to provide reliable measurements of the much lower concentrations found in non-industrial

environments. A size selective sampler is needed for collecting inhalable particles ($d < 10 \mu\text{m}$) in a variety of environments over sampling times of 1 to 4 hours. While direct-reading instruments exist that permit 2 to 5 minute integrated data, no physical sample is collected. A particulate sampler should be evaluated (for cutoff size, efficiency, and air velocity effects) with reference to other conventional samplers. Ideally, samplers should be suitable for both fixed locations and personal monitoring. The National Bureau of Standards stack-filter sampler may be appropriate after independent testing. However, the use of tandem stacked filters to achieve size separation requires further evaluations under both laboratory and field conditions.

(6) *Tobacco Smoke*. Tobacco-specific nitrosamines have been studied by several groups (Brunnemann *et al.*, 1977, 1978, 1980; Hoffman *et al.*, 1979b; Stehlik *et al.*, 1982; Matsushita *et al.*, 1983). American cigarettes contain 300 to 500 ng of *N*-nitroso-dimethylamine and 100–300 ng of *N*-nitrosopyrrolidine (Matsushita and Mori, 1984). Air samples are analysed by extraction and clean-up, followed by gas/liquid chromatography with a thermal energy analyser detector which detects the nitrosamines by pyrolysis, cleaving the N-N bonds and measuring the resulting nitric oxide by chemiluminescence (Jenkins and Guerin, 1984). Tobacco smoke has been demonstrated to be an irritant, and is possibly a contributor to respiratory illness. To date, epidemiological studies have not incorporated direct exposure measurements of tobacco smoke. Stable indicators of tobacco-smoke components of indoor suspended particulate matter are needed. *N*-nitrosodimethylamine and *N*-nitrosopyrrolidine appear to be promising indoor tracers. Nicotine, a unique component of tobacco smoke, also serves as a tracer. In intercomparisons of both active and passive methods, nicotine samplers have been demonstrated to be reliable. Further, greater than 95% of the nicotine is in the gas phase (Caka *et al.*, 1990). However, since nicotine can exist in both particulate and vapour phases at room temperature, care must be taken to quantify each component.

(7) *Organic Compound Determination*. Volatile and nonvolatile organic compounds are ubiquitous indoors. Because of the numerous consumer products containing organic chemicals, indoor concentrations are usually higher than outdoors. Within-home and among-home variability of two orders of magnitude have been reported. Several sampling media are currently available, including Tenax, polyurethane foam, molecular sieves, and charcoal. For each of these, sample recovery efficiency, breakthrough volumes, intersample variability, sample stability with temperature and humidity, and artifact formations need to be determined.

(8) *Passive Formaldehyde Sampler*. An inexpensive passive formaldehyde monitor is needed. This device should be capable of detecting 0.02 ppm over a 1 to 4 hour exposure period. Such a device would find widespread application in homes and offices. Because the irritant properties of formaldehyde are commonly associated with sources only in distinct identifiable locations, a personal monitoring device for formaldehyde would not appear to be required.

6 REFERENCES

- Atherholt, T.B., *et al.* (1985). Mutagenicity studies of New Jersey air particulate extracts. In Waters, M.D. *et al.* (Eds.) *Short-Term Bioassays in the Analysis of Complex Environmental Mixtures, IV*, pp. 211–231. Plenum Press, New York.
- Bright, D.S. and Fletcher, R.A. (1983). New portable ambient aerosol sampler. *Am. Ind. Hyg. Assoc. J.* **44**, 528–536.
- Brunnemann, K.D. and Hoffman, D. (1978). In *Chemical studies on tobacco smoke*. LIX. Analysis of volatile nitrosamines in tobacco smoke and polluted indoor environments. ARC Sci. Publ. 19, ARC Press, New York.
- Brunnemann, K.D., Yu, L., and Hoffman, D. (1977). Assessment of carcinogenic volatile N-nitrosamines in tobacco and in mainstream and sidestream smoke from cigarettes. *Cancer Res.* **37**, 3218–3222.
- Brunnemann, K.D., Fink, W., and Moser, F. (1980). Analysis of volatile N-nitrosamines in mainstream and sidestream smoke from cigarettes by GLC-TEA. *Oncology* **37**, 217–222.
- Butler, J.P., Kneip, T.J., and Daisey, J.M. (1987). An investigation of interurban variation in the chemical composition and mutagenic activity of airborne particulate organic matter using an integrated chemical glass/bioassay system. *Atmos. Environ.* **21**, 883–892.
- Caka, F.M., *et al.* (1990). An intercomparison of sampling techniques for nicotine in indoor environments. *Environ. Sci. Technol.* **24**, 1196–1203.
- Colome, S.D., Spengler, J.D., and McCarthy, S. (1982). Characterisation of aerosols and inorganic gases in indoor environments. *Environ. Int.*, **8**, 197–204.
- Comstock, G.W., Meyer, M.B., Helsing, K.F., and Tockman, M.S. (1981). Respiratory effects of household exposures to tobacco smoke and gas cooking. *Am. Rev. Respir. Dis.* **124**, 143–148.
- Daley, P.S. and Lundgren, D.A. (1975). The performance of piezoelectric crystal sensors used to determine aerosol mean concentrations. *Am. Ind. Hyg. Assoc. J.* **36**, 518–524.
- de Bortoli, M., *et al.* (1984). Integrated “real life” measurements of organic pollution in indoor and outdoor air of homes in Northern Italy. *Proceedings of the 3rd International Conference on Indoor Air Quality and Climate*, Stockholm, August 20–24, 1984.
- Dockery, D.W. and Spengler, J.D. (1981). Personal exposure to respirable particulates and sulfates. *J. Air Pollut. Control Assoc.* **31**, 153.
- Fletcher, R.A. (1984). A review of personal/portable monitors and samplers for airborne particles. *J. Air Pollut. Control Assoc.* **34**, 1014–1016.
- Forlenza, V.A. (1983). Laboratory studies of a passive electrochemical instrument for measuring carbon monoxide. In *Proceedings: National Symposium on Recent Advances in Pollutant Monitoring of Ambient Air and Stationary Sources.*, pp. 358–368. Report No. EPA 600/9–83–007. U.S. Environmental Protection Agency, Research Triangle Park, North Carolina.
- Goldstein, B.D., *et al.* (1979). The relation between respiratory illness in primary school children and the use of gas for cooking. II. Factors affecting nitrogen dioxide levels in the home. *Int. J. Epidemiol.* **8**, 339–345.
- Good, B.W., Vilcins, G., Harvey, W.R., Clabo, D.A., and Lewis, A.L. (1982). Effect of cigarette smoking on residential NO₂ levels. *Environ. Int.* **8**, 167–175.
- Grimmer, G., Bohnke, H., and Harke, H.P. (1977). Passive smoking: measuring concentration of polycyclic aromatic hydrocarbons in rooms after machine smoking of cigarettes. *Int. Arch. Occup. Environ. Health* **40**, 83–92.
- Gruber, A.H., Goldstein, A.G., La Conti, A.B., Wheeler, H.G., and Martin, J. (1979). A new family of miniaturised self-contained CO dosimeters and direct reading detectors. *Proceedings of the Symposium on the Development and Usage of Personal Monitors for Exposure and Health Effect Studies*, pp. 83–99. Report

- No. EPA 600/9-79-032, U.S. Environmental Protection Agency, Research Triangle Park, North Carolina.
- Harkov, R., Greenberg, A., Darack F., Daisey, J.M., and Liroy, P.J. (1984). Summertime variations in polycyclic aromatic hydrocarbons at four sites in New Jersey. *Environ. Sci. Technol.* **18**, 287-291.
- Hartwell, T.P., et al. (1984). *Study of Carbon Monoxide Exposure of Residents of Washington D.C. and Denver, Colorado, Parts I and II*. Final Report. Contract £68-02-3679. U.S. Environmental Protection Agency, Research Triangle Park, North Carolina.
- Hoffman, D., Adams, J.D., and Wynder, E.L. (1979a). Formation and analysis of carbon monoxide in cigarette mainstream and sidestream smoke. *Prev. Med.* **8**, 344-350.
- Hoffman, D., Adams, J., Brunnemann, K.D., and Hecht, S.S. (1979b). Assessment of tobacco-specific N-nitrosamines in tobacco products. *Cancer Res.* **39**, 2505-2509.
- Hollowell, C. and Miksch, R. (1981). Sources and concentrations of organic compounds in indoor environments. *Bull. NY Acad. Med.* **57**, 962-977.
- Immerman, F.W. and J.L. Schaum (1990). *Nonoccupational Pesticide Exposure Study (NOPES)*. Project Summary EPA 600/S3-90/003. U.S. Environmental Protection Agency, Research Triangle Park, North Carolina.
- Jenkins, R.A. and Guerin, M.R. (1984). Analytical chemical methods for the detection of environmental tobacco smoke constituents. *Eur. J. Respir. Dis. Suppl.* **133**, 33-46.
- Johnson, T. (1983). *A Study of Personal Exposure to Carbon Monoxide in Denver, Colorado*. Final Report. Contract £68-02-3755. U.S. Environmental Protection Agency, Research Triangle Park, North Carolina.
- Kosek, J.A., Giordano, J.P., and La Conti, A.B. (1983). Development of solid polymer electrolyte head instrumentation. In *Proceedings of the National Symposium on Recent Advances in Pollutant Monitoring of Ambient Air and Stationary Sources*, pp. 333-357. Report No. EPA 600/9-83-007. U.S. Environmental Protection Agency, Research Triangle Park, North Carolina.
- Krause, C., et al. (1987). Occurrence of volatile organic compounds in the air of 500 homes in the Federal Republic of Germany. In *Proceedings of The 4th International Conference on Indoor Air Quality and Climate*, Vol. 1, pp. 102-106. Institute for Soil, Water, and Air Hygiene, Berlin.
- Lebret, E., Van de Weil, H.J., Noij, D., and Boleij, J.S.M. (1986). Volatile hydrocarbons in Dutch homes. *Environ. Int.* **12** (1-4), 323-332.
- Liroy, P.J., et al. (1983). The New Jersey project on airborne toxic elements and organic species (ATEOS). *JAPCA* **33**, 649-657.
- Liroy, P.J., Avdenko, M.A., Harkov, R., Atherholt, T., and Daisey, J.M. (1985). A pilot indoor-outdoor study of organic particulate matter and particulate mutagenicity. *JAPCA* **35**, 653-657.
- Matsushita, H. and Mori, T. (1984). Nitrogen dioxide and nitrosamine levels in indoor air and side-stream smoke from cigarettes and its application to Japanese and foreign cigarettes. *J. Japan Soc. Air Pollut.* **18**, 339.
- Matsushita, H., Mori, T., and Goto, S. (1983). An improved method for analysing N-nitrosamines in sidestream smoke from cigarettes and its application to Japanese and foreign cigarettes. *J. Japan Soc. Air Pollut.* **18**, 339.
- Melia, R.J.W., Florey, C. duV., Altman, D.G., and Swan, A.V. (1977). Association between gas cooking and respiratory disease in children. *Br. Med. J.* **2**, 149-152.
- Melia, R.J.W., et al. (1982). Childhood respiratory illness and the home environment. II. Association between respiratory illness and nitrogen dioxide, temperature, and relative humidity. *Int. J. Epidemiol.* **11**, 164-169.
- Morandi, M.T., Daisey, J.M. and Liroy, P.J. (1987). Development of a modified factor analysis/multiple regression model to apportion suspended particulate matter in a complex airshed. *Atmos. Environ.* **21**, 1821-1829.

- Nitta, H. and Maeda, K. (1982). Personal exposure monitoring to nitrogen dioxide. *Environ. Int.* **8**, 243-248.
- Ott, W.R. (1990). Total human exposure: basic concepts, EPA field studies, and future research needs. *J. Air Waste Management Assoc.* **40**(7), 966-975.
- Ott, W., Williams, C., Rhodes, C., Drago, R., and Burmann, F. (1984). Application of Microprocessors to Data Logging Problems in Air. Paper presented at the 77th annual meeting of the Air Pollution Control Association, San Francisco, California, June 27-29, 1984.
- Palmer, E.D., Gunnison, A.F., Dimattio, J., and Tomczyk, C. (1976). Personal sampler for nitrogen dioxide. *Am. Ind. Hyg. Assoc. J.* **37**, 570-577.
- Pellizzari, E.D., et al. (1984a). *Total Exposure Assessment Methodology (TEAM) Study: First Season, Northern New Jersey*. Contract No. 6802-3679. U.S. Environmental Protection Agency, Office of Research and Development, Washington, D.C.
- Pellizzari, E.D., et al. (1984b). Sampling and analysis for volatile organics in indoor and outdoor air in New Jersey. In *Indoor Air*, Vol. 4, pp. 221-226. Swedish Council for Building Research, Stockholm.
- Pellizzari, E.D., et al. (1984c). Volatile organic levels in indoor air. In *Indoor Air* Vol. 4, pp. 303-308. Swedish Council for Building Research, Stockholm.
- Quant, R.R., Nelson, P.A., and Sem, G.J. (1982). Experimental measurements of aerosol concentrations in offices. *Environ. Int.* **8**, 223-227.
- Repace, J. and Lowrey, A. (1980). Indoor air pollution, tobacco smoke, and public health. *Science* **208**, 464.
- Rounbehler, D.P., Reich, S., Coombs, J., and Fine, D.H. (1980). Nitrosamine air sampling sorbents compared for quantitative collection and artifact formation. *Anal. Chem.* **52**, pp. 273-276.
- Sem, G.J. and Quant, F.R. (1983). Automatic piezobalance respirable aerosol mass monitor for unattended real-time measurements. In Marple, V.A. and Liu, B.Y.H. (Eds.) *Aerosols in the Mining and Industrial Work Environments*, Vol. 3, *Instrumentation*, Ann Arbor Science Publ, Ann Arbor, Michigan.
- Sem, G.J. and Tsurubayashi, K. (1975). A new sensor for respirable dust measurements. *Am. Ind. Hyg. Assoc.* **36**, 791-800.
- Sem, G.J., Tsurubayashi, K., and Homnia, K. (1977). Performance of the piezoelectric microbalance respirable aerosol sensor. *Am. Ind. Hyg. Assoc. J.* **38**, 580-586.
- Sheldon, L.S., et al. (1988a). *Indoor Air Quality in Public Buildings: Vol. I*. EPA 600/6-88/009a. NTIS PB 89-102503/AS. U.S. Environmental Protection Agency, Washington, DC.
- Sheldon, L.S., Zelon, H.S., Sickles, J., Eaton, C. and Hartwell, T. (1988b). *Indoor Air Quality in Public Buildings: Vol. II*. EPA 600/6-88/009b. NTIS PB 89-102511/AS. U.S. Environmental Protection Agency, Research Triangle Park, North Carolina.
- Spengler, J.D. and Soczek, M.L. (1984). Evidence for improved ambient air quality and the need for personal exposure research. *Environ. Sci. Technol.* **18**, 268A-280A.
- Spengler, J.D., Duffy, C.P., Letz, R., Tibbits, T., and Ferris, B.G. (1983). Nitrogen dioxide inside and outside 137 homes and implications for ambient air quality standards and health effects research. *Environ. Sci. Technol.* **17**, 114-168.
- Stehlik, G., Richter, O., and Altmann, H. (1982). Concentration of dimethylnitrosamine in the air of smoke-filled rooms. *Ecotoxicol. Environ. Safety* **6**, 495-500.
- Stetter, J.R., Rutt, D.R., and Graves, M.R. (1979). Electrochemical methods for development of personal exposure monitors. *Proceedings of the Symposium on the Development and Usage of Personal Monitors for Exposure and Health Effect Studies*, pp. 35-46. Report No. EPA 600/9-79-032. U.S. Environmental Protection Agency, Research Triangle Park, North Carolina.

- Stevens, R.K. (1984). Sampling and Analysis of Atmospheric Aerosols. Paper presented at the Advanced Research Workshop on Environmental Monitoring of Architectural Conservation, Rome, Italy, June 11–14, 1984.
- Turner, W.A., Marple, V.A. and Spengler, J.D. (1984). Indoor Aerosol Impactor. Paper presented at the AAAR meeting, Minneapolis, Minnesota, September 1984. American Association for Aerosol Research, Research Triangle Park, North Carolina.
- Turner, W.A., Spengler, J.D., Dockery, D.W., and Colome, S.D. (1979). Design and performance of a reliable personal monitoring system for respirable particulates. *J. Air Pollut. Control Assoc.* **29**, 747.
- Vo-Dinh, T. and Hooyman, J.R. (1979). Selective heavy atom perturbation for analysis of complex mixtures by room temperature phosphorimetry. *Anal. Chem.* **51**, 1915.
- Vo-Dinh, T. and Winefordern, J.D. (1977). Room temperature phosphorimetry as a new spectrochemical method of analysis. *Appl. Spectrosc. Rev.* **13**, 261.
- Vo-Dinh, T., Gammage, R.B., and Martinez, P.R. (1980). Identification and quantification of polynuclear aromatic compounds in synthoil by room temperature phosphorimetry. *Anal. Chim. Acta* **118**, 313.
- Vo-Dinh, T., Gammage, R.B., and Martinez, P.R. (1981). Analysis of a workplace air particulate sample by synchronous luminescence and room temperature phosphorescence. *Anal. Chem.* **53**, 253–258.
- Wallace, L.A. (1987). Total exposure assessment methodology (TEAM) study: Summary and analysis, Vol. 1. United States Environmental Protection Agency, Washington, DC.
- Wallace, L.A. *et al.* (1988). The California TEAM Study: Breath concentrations and personal exposures to 26 volatile compounds in air and drinking water of 188 residents of Los Angeles, Antioch, and Pittsburgh, CA. *Atmos. Environ.* **22**, 2141–2163.
- Wallace, L.A., Thomas, J., and Mage, D. (1984). Alveolar CO Measurements of 1000 Residents of Denver and Washington, D.C.—Comparison with Preceding Personal Exposures. Paper 84–121.5 presented at the annual meeting of the Air Pollution Control Association, San Francisco, California, June 27–29, 1984.
- Wallace, L.A., Pellizzari, E.D., and Gordon, S.M. (1985a). Organic chemicals in indoor air: A review of human exposure studies and indoor air quality studies. In *Proceedings of the 7th Annual ORNL Life Sciences Symposium*, Oak Ridge National Laboratory, Oak Ridge, Tennessee.
- Wallace, L.A., Pellizzari, E.D., Hartwell, T., Sparacino, C., and Zelon, H. (1985b). Personal exposures, indoor-outdoor relationships, and breath levels of volatile organics in New Jersey. *Atmos. Environ.* **19**, 1651–1661.
- Yanagisawa, Y. and Nishimura, H. (1982). Badge-type personal sampler for measurement of personal exposures to NO₂ and NO in ambient air. *Environ. Int.* **8**, 235–242.