
2 Exposure Measurements*

This chapter is concerned with the methods of quantifying the amount of a chemical contaminant that has the potential of reacting with a given receptor. A receptor is broadly defined to include living organisms or a non-living entity such as decorative statues. We are concerned with contaminants that, in sufficient quantities or duration, are suspected to cause or to contribute to short-term or long-term adverse effects. These effects may or may not be reversible. In dealing with environmental effects, it is important to note that, given the heterogeneity in response within species, an individual member of a species may be irreversibly harmed without jeopardising the species as a whole, or the ecosystem. Such distinctions are not usually acceptable with regard to human health effects.

There is a broad range of contaminants whose chemical or physical properties may have effects in concentrations likely to be present in the environment. These materials may be either naturally-occurring, such as aflatoxins, or produced as a primary or secondary result of human activities. For instance, ozone occurs naturally in the upper atmosphere but also is produced by photochemical reactions among precursors generated in fossil fuel combustion and by evaporation of hydrocarbons, and by electrical ionisation of air (high tension lines, motors). Excluded, however, from this report are biological agents (virus, bacteria, spores, pollen), noise, and ionising and non-ionising radiation. This context does not exclude the use of biological materials or even organisms as measures of exposure.

Before proceeding to discuss exposure measurement methodologies, it is necessary to discuss the topic boundaries of this chapter. One can imagine a risk model for environmental pollution as comprised of five distinct components:

- (1) source(s) of pollutants,
- (2) transport of these pollutants from sources to target organisms,
- (3) exposures of a target organism to these pollutants,
- (4) doses received by the organisms who are thereby exposed, and
- (5) effects resulting from these exposures.

These five components may be viewed as links in a chain: the output of each component is the input to the next. Each component may be separated and studied independently; for example, if it is found that a target population

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experiences zero exposure to a given pollutant, then it is not necessary to study the other four components, because no effects are possible. Of the five, the two for which the least information is available on most pollutants are exposure and dose. This chapter is concerned only with the third component: determining the exposures of a population of interest. Before the methodology for determining exposures can be discussed, it is necessary to define what is meant by an "exposure."

2.1 DEFINITION OF EXPOSURE

To define exposure it is necessary to specify who is exposed to what substance. Too often, one or the other component is only loosely defined or understood, thereby resulting in confusion and disagreement. The target for exposure may be an intact organism, a particular organ, or even a particular cellular component. Exposure occurs when the pollutant is present at the boundary of the target. To complete the definition, it may also be necessary to specify the physico-chemical form of the pollutant of interest. The boundary at the target may not necessarily be physically tangible, but should correspond with the conceptual interface between what is inside the organism and what is outside. Three examples illustrate this concept. If a person is present in air containing 20 parts per million (ppm) of carbon monoxide (CO) at 8:30 AM, then we say that the person is exposed to 20 ppm CO at that instant. Similarly, if a mussel is present in waters containing $85 \mu\text{g}/\text{m}^3$ of cadmium (Cd) at 4:00 PM on a given date, then we say that the mussel is exposed to $85 \mu\text{g}/\text{m}^3$ Cd at that time and date. When material that is drunk or eaten is being considered, exposure occurs only when that material is presented for ingestion. In all cases, the pollutant is present at the same location in space as the conceptual or physical boundary at the target and at the same time.

The target is exposed because it comes into contact with the pollutant. If the pollutant transfers across the boundary and enters the space occupied by the target, a dose occurs. Thus there can be exposure without a dose but not a dose without exposure. In many cases physico-chemical or biological models may be used to convert the calculated dose for the target into a dose for the particular organ or site within the target; for example the human kidney or the central nervous system of a bird of prey. A formal conceptual framework for exposure and dose has been prepared for air pollutants and humans (Ott, 1982), and this conceptual framework can be extended to other environmental media and to nonhuman forms. The following definition is a generalisation of earlier definitions in the literature (Vouk, *et al.* 1985; Ott, 1982, 1984).

Imagine a three-dimensional envelope enclosing the organism (see Figure 2.1), and assume this envelope is located at some position (coordinates x, y, z) in space at time t . For a human being, the location of person i at time t can be expressed as (x, y, z, t) , where x, y , and z are distances from the origin in some urban reference system. Since pollutants vary in time and space, the concentration present in space at time t also can be described using this same reference system: $c(x, y, z, t)$. Then, an exposure of person i to concentration c occurs when the following events occur jointly:

person i is present at location x, y, z at time t ;

concentration c is present at location x, y, z at time t .

Thus an "exposure" may be defined as an event occurring at some location.

Although many monitoring instruments can generate "continuous" readings giving the concentration at a particular location at almost any instantaneous time, many pollutants can be monitored only by techniques that integrate the concentration over some minimum time T . That is, they provide the integrated exposure of organism i located at position (x, y, z) :

$$E_i(T) = \int_0^T c(x, y, z, t) dt \quad (1)$$

When the integrated exposure $E_i(T)$ is divided by the time T , the result

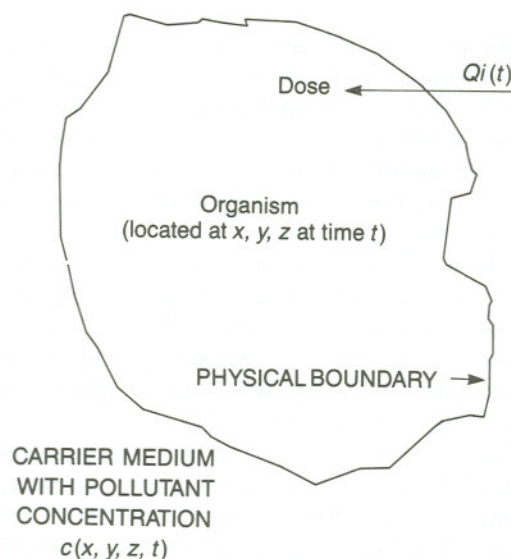


Figure 2.1. Graphical representation of the exposure and dosage received by organism i at location (x, y, z) and time t to a pollutant in a carrier medium with concentration $c(x, y, z, t)$

is the average rate of exposure of person i , which is useful for many environmental applications:

$$AE_i(T) = \frac{E_i(T)}{T} \quad (2)$$

Usually, monitoring instruments and methods have inherent limitations which make it impossible to obtain instantaneous readings of concentration. When the minimum averaging time T that is possible with a given measurement method is much greater than the time required to cause adverse effects on the organism, then the relevance of such data for estimating adverse effects on the organism is questionable.

Adequate exposure measurement methods with suitable time resolution exist for some environmental pollutants. For those measurement methods with insufficient time resolution, it is recommended that improved exposure measurement methods be developed.

2.2 OBJECTIVES OF EXPOSURE ASSESSMENT

There are multiple purposes for assessing exposures to contaminants in the environment. Assessment strategies should be devised in response to the objectives or to the particular hypotheses being tested. In turn, this will define the contaminant(s), their physico-chemical form, and the population to be studied. Subsequently, decisions on instrumentation, analytical and statistical methods, sample size, survey methods, location, frequency, timing and duration of measurements can be made.

In terms of biological hypotheses for studies of health and environmental effects, distinctions between fluctuating concentrations (and possibly short-term peaks) versus integrated exposures will have important implications for methods and for instrument requirements. In addition, the possibility of interactive or target-specific factors will influence the selection of ancillary components of exposure assessment studies.

Exposure assessment can be categorised in three non-exclusionary general objectives (Table 2.1). Exploratory assessments are useful to provide a general understanding of the intensity and diversity of exposures occurring within a population. Improving the understanding of health and ecological effects is a possible outcome from quantification of exposures. Finally, the regulatory process, in its broad sense, becomes more cost-effective with better information on contaminant exposures within and among media.

Table 2.1. Objectives of exposure assessment

(1) Exploration
– Define the biological questions of concern
– Define the population distribution to a contaminant
– Determine the comparative contribution to exposures by activity, location, source
– Provide a basis for cross-culture comparisons
– Provide a basis for cross-media comparisons
(2) Definition of Health/Ecological Impacts
– Integrate health questions or measurement with exposure metrics to be applied.
– Establish variability as a basis to determine sample size of study populations
– Determine sub-populations at higher risk due to greater exposure
– Determine relative risks by media and by contaminant
– Quantify exposure/response function with measured uncertainty
(3) Regulation
– Determine the relative source contributions for developing strategies to decrease exposure
– Ascertain and document trends
– Monitor compliance and determine effectiveness of implemented strategies

2.3 APPROACHES FOR DETERMINING EXPOSURES

Accurate measurement of one member of a population still leaves several questions concerning the population as a whole: “What are the exposures of other members of the population?” “How do exposures vary from organism to organism?” “What sources are responsible for these exposures?”

To characterise statistically the exposures of a population, a single point estimate (the mean, median, mode, etc.) is insufficient. Because adverse effects usually are associated with exposure to the highest concentrations of a chemical, we are mostly interested in determining the percentage of the population exposed to these concentrations. Ideally the frequency distribution of exposures of a population over space and time is needed (Figure 2.2). From such a frequency distribution, it is possible to determine the proportion of a population exposed to most concentrations. For example, if it is determined that 99% of the population should be below a maximum permissible limit, then the degree of source reduction needed to obtain this goal can be calculated.

There are two conceptually different approaches for determining a frequency distribution of exposure: the direct and indirect approaches (Duan, 1982). We give a brief discussion of the two approaches below.

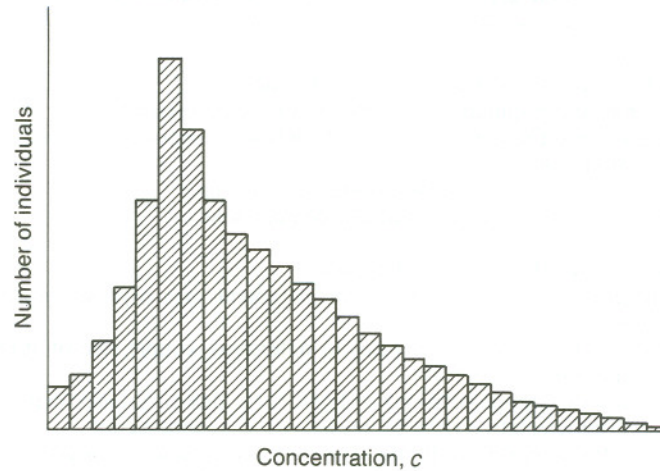


Figure 2.2. Hypothetical frequency distribution of the exposures of a population over space and time

2.3.1 DIRECT APPROACH

Obtaining an exposure measurement of every member of the population to characterise the frequency distribution of exposures usually is impossible. However, if a measurement method is capable of measuring an individual organism's exposure, it may be possible to use this technique on a group of organisms.

In one approach, individuals are selected according to the well-established procedures of probability sampling, then the resulting frequency distribution provides a good approximation of the frequency distribution of exposures of the entire population. When individuals suspected of having unusually high exposures are purposely included in greater proportions than other individuals, the approach is called "stratification", and the result is called a stratified random sample.

2.3.2 INDIRECT APPROACH

If it is impossible to use the direct approach to obtain the exposure frequency distribution, then it is necessary to estimate this distribution by another method. If the organism is mobile and known to relocate to other micro-environments, the exposure distribution is estimated from the concentration in each micro-environment and the time of occupancy. If, for example, c_j is the concentration in micro-environment j and t_{ij} is the time spent by organism i in micro-environment j , the integrated exposure E_i for the organism is computed as follows:

$$E_i = \sum_{j=1}^n c_{ij} t_{ij} \quad (3)$$

Dividing by an averaging time t , the average exposure of organism i is obtained for the averaging period. If a similar computation is performed for every member of the population, then the resulting exposures can be assembled as an estimated frequency distribution of exposures of the population.

For a material that is drunk or eaten, exposure occurs only when that material is presented for ingestion. In all cases, the pollutant is present at the same location in space as the conceptual or physical interchange surface of the target, and at the same time; the time of the event is instantaneous.

The target is exposed when it comes into contact with the pollutant through the interchange surfaces. If the pollutant transfers across the boundary and enters the space occupied by the target, a dose is delivered. Thus there can be exposure without a dose, but not a dose without exposure. In many cases physico-chemical or biological models may be used to convert the calculated dose for the whole body into the exposure and the dose for the particular organ or site within the target organ, for example, the human kidney or the central nervous system of a bird of prey. This definition offers the flexibility that both whole body exposure and exposure to internal organs can be considered within the same framework.

When the integrated exposure $E_i(0,t)$ is divided by the time T , the result will be the average exposure of person i :

$$AE_i(0,t) = \frac{E_i(0,t)}{T} \quad (4)$$

2.4 STUDY DESIGNS

2.4.1 DIRECT APPROACH

There are two basic approaches for determining exposure of a population. One is called the direct approach (Duan, 1982) where the exposure profiles of a sample of persons are combined for extrapolation to the entire population. The second approach, called indirect, develops simulated exposure profiles by combining information on the time people spend in particular activities with the concentrations associated with those activities.

The direct approach described in this section is truly a new methodology applied to several types of pollutants only in the last five years (Akland *et al.*, 1985; Hartwell *et al.*, 1984; Spengler *et al.*, 1985). The uniqueness of the direct approach is that it combines the survey research techniques of the social scientist with the measurement technology of the chemist. The

main component of the direct approach is a field study utilising survey research methodology and environmental monitoring with personal exposure monitors. Whereas there is conclusive evidence that survey research methodologies are successful in many applications, problems have been identified which are discussed in this section.

While personal exposure monitoring devices have been well documented in the literature (Wallace and Ott, 1982), there has been very little written about survey methods in human air pollution exposure field studies (Croce *et al.*, 1985).

The field study survey design consists of some basic components briefly described below:

- (1) Appropriateness of the survey instrument to answer the questions under investigation;
- (2) An Analysis Plan to describe how the data from the field study are to be tabulated and analysed;
- (3) A Questionnaire or structured set of questions to the respondents in the study;
- (4) The Interviewing Strategy, the method by which the respondents are to be asked questions and to receive personal monitors along with use instructions;
- (5) The Sampling Plan is the protocol to determine how many members of the target population should be in a sample and how individuals are to be selected.

Probability sampling methods are commonly used in field studies to allow inferences to the entire target population. The direct approach presents a practical and definable way to assess exposure profiles of a target population. An important and unique feature of this approach is that a profile can be developed with a known level of precision based on probability sampling methods used. Survey techniques can be adapted easily and inexpensively for use in other studies. The survey methodology is useful not only for the direct approach in media other than air, but also is applicable to the indirect approach (Duan, 1985). For food, the method of choice for direct dietary exposure measurements is the double portion study. However, in spite of the successful application of the direct method in recent years, there is a need to improve survey methodology.

2.4.1.1 Improvement of Survey Response Rate in the Direct Approach

Poor compliance in surveys can be another significant source of bias, often due to failure of respondents either to follow instructions about the placement or use of the instruments or to answer specific questions in a questionnaire or diary. It appears that, even though respondents agree to participate in a field study, they might not be able to do so effectively for any number of

reasons, such as inadequate or unclear instructions; inability to follow instructions; lack of incentive or desire to participate beyond a point; lack of clarity or understanding of specific questions; or lack of information needed to respond to specific questions.

Some of these reasons relate to the survey designers' failure to understand human behaviour. Other reasons relate to poor design. The failure to better understand human behaviour in regard to environmental monitoring at this time can be remedied by conducting methodological investigations primarily employing focus group interviews involving the assistance of behavioural scientists.

A significant source of bias is an incomplete response rate, which will adversely affect population estimates if non-responders differ from responders in their exposure profiles. When probability sampling techniques are used to select subjects to be monitored, weight adjustments based on response can be made to compensate for potential bias due to non-response, using specified assumptions about the nature of the non-response. However, if those assumptions are incorrect, the estimates might still be biased, despite such weight adjustments. Survey statisticians generally recommend a 75 percent response rate to minimise such risk of bias (Croce *et al.*, 1985).

Methodological investigations to achieve this goal should cover the following issues: (1) how respondent cooperation is solicited in general; (2) the amount of time and effort designated respondents are required to devote to a survey; and (3) the effectiveness of incentives for participants. Several comments need to be made in regard to these three issues.

How an individual is approached and asked to participate is extremely critical. What is said and who makes the presentation both influence the outcome.

Since participants are asked to devote a specific amount of time, there can be difficulties arising from individuals being too busy to participate, having competing commitments, or being simply unwilling to participate.

Generally, respondents can be encouraged to participate through the payment of incentives; for instance, respondents in the USA are paid cash in return for their participation. The importance of incentives in stimulating respondent cooperation in a field study is generally unknown, however. Of particular interest is the need to assess the relative value of cash versus non-monetary incentives, and the relative effectiveness of paying a constant sum or variable sum of money depending on the socio-economic status of each study participant.

2.4.2 STUDY DESIGN: INDIRECT APPROACH FOR HUMAN EXPOSURE

As defined above, the integrated exposure for an organism to a chemical in a given time period (which is usually the objective of the exposure study)

is the accumulation of the concentrations experienced at different instances in different components of the environment. In the direct approach used for air pollutants, it is possible to have samplers continually measuring the concentrations of chemicals in inhaled air. Analogous procedures can be envisaged to determine human exposures via food, for example, by duplicate diet sampling, and via water consumption using an appropriate sampling device. Such direct approaches, if feasible, are preferred.

To assess exposure of non-human biota, and in some cases that of humans, the direct approach is not always feasible because of instrumental or economic limitations. In these cases, exposure must be assessed indirectly by measuring concentrations of chemicals in different environmental samples taken at different times and locations. Total exposure is then assessed by combining the contributions of these micro-environmental measurements.

The indirect approach is discussed in further detail below. The application of this approach to human exposure to chemicals in air is addressed first, followed by that for human exposure by ingestion, and finally by that for non-human exposure. Further details on the conceptual framework of the indirect approach for air pollutants and some empirical results are given in Duan (1985).

2.4.2.1 Human Exposure to Air Pollution

The indirect approach is distinguished from the direct one by the reconstitution of the integrated exposure according to the micro-environment types (METs). A micro-environment is defined as a segment of air space (or of space/time) with homogeneous pollutant concentrations. An MET is a group of similar micro-environments. For example, all indoor micro-environments might be grouped together as the indoor MET, and all outdoor micro-environments together as the outdoor MET. In terms of the MET's, integrated exposures are reformulated as follows:

$$E_i = \sum_{k=1}^n C_{ik} T_{ik} \quad (5)$$

Where E_i is the integrated exposure of the i th observation unit, C_{ik} is the average concentration confronted by the i th observation unit in the k th MET, and T_{ik} is the amount of time the i th observation unit spends in the micro-environment. For particulates which vary in size, we need to construct a size profile using several ranges of discrete sizes with each size range addressed separately.

The indirect approach depends crucially on the assumption that MET concentrations C are independent of MET time allocation T . Under this assumption, it is possible to use concentration data and time allocation data from two different sources and to combine the two analytically to produce

integrated exposures. First, design considerations for obtaining concentration and time-allocation data are discussed; subsequently, the analytical methods to combine concentration and time-allocation data to estimate integrated exposure are evaluated.

Design Time allocation data must be obtained from a probabilistic sample of human subjects. The methodologies are the same as those discussed in the section on the direct approach, with the exception that monitoring instruments are replaced by survey instruments such as activity diaries or a recall survey. Since the survey instruments are likely to be more convenient to the human subjects than the monitoring instruments, cooperation is likely to be improved.

Concentration data can be collected in two ways. The first approach involves personal monitoring, referred to as the enhanced personal monitoring (EPM) approach. The second involves micro-environment monitoring (MEM); specifically, a sample of micro-environments is monitored for each MET.

With EPM, MET concentration data are collected on a probabilistic sample of human subjects, and time-allocation data on another probabilistic sample of human subjects.

The monitoring phase of an EPM study can be more demanding than the monitoring in the direct approach. With the former, the MET in which exposure occurs must be identified; this will typically require monitoring instruments capable of giving reliable continuous measurements. With the latter, only the measurement of integrated exposure is needed, and can be implemented using badge type passive dosimeters.

The probabilistic samples of human subjects—one for monitoring, one for time allocation—can either overlap, or be disjointed. For example, in a personal monitoring study, additional use can be made of the data by supplementing the monitored sample with an adjunct sample on which only time-allocation data are collected. In this case, the two samples overlap—the monitoring sample being a subset of the second or time-allocation sample. Furthermore, the update of a previous personal monitoring study by collecting new time-allocation data may be desirable. Assuming that the MET concentration data in the previous study remain representative of the current population, two samples exist: the monitoring sample from the previous study and the time-allocation sample from the new one.

With MEM, a number of micro-environments are sampled from each MET, and those micro-environments are monitored directly. The results are then combined with MET time-allocation data collected from a probabilistic sample of human subjects. The main advantage of MEM is the lack of need to monitor individual subjects. Actually, MEM does not require personal monitors—portable or mobile monitors can be used as well, because the monitoring is performed solely by trained technicians.

The sampling of micro-environments poses difficulties remaining to be

studied. Note that the target population is not that of micro-environments belonging to a certain MET. Consider, for example, the MET consisting of all office spaces, and assume that each identifiable room is sufficiently homogeneous and can be regarded as a micro-environment. If the collection of all micro-environments were the target population, a roster of all office spaces could be the sampling frame. However, the appropriate target populations should be the occupants of the micro-environment. Some office spaces might be empty most of the time, some might be crowded most of the time. To obtain a sample of office spaces representative of concentrations confronted by humans, adjustments for such differences are needed. Two preliminary proposals for the sampling of micro-environments are described in Duan (1985): the weighted sampling scheme and the simulated human activities. These proposals remain to be validated in field work.

Analysis A statistical model which underlies the indirect approach to estimate exposure using MET concentration and time-allocation data is a primary consideration. The term "model" is used in its broadest sense; the only modeling assumptions required are statistical ones related to properties of concentration and time-allocation, namely, (1) that the MET concentration C and time allocations T are independent, (2) that the observed MET concentrations are representative of the MET concentrations in the target population, and (3) that the observed time-allocations are representative of the broader situation. These assumptions are substantially weaker than the physical/biological assumptions in transport/uptake models, the details of which are given in the later part of this volume.

With these three assumptions, two general approaches to estimate exposure are evident. One uses simulation models such as SHAPE. This approach summarises the observed concentration and time-allocation data by parametric or non-parametric probability (frequency) distributions, generates hypothetical concentration and time-allocation data from these distributions, and inputs the resulting exposures and exposure distributions. Several comments are particularly relevant:

(1) Simulation models are useful for filling gaps in empirical data. For example, in earlier versions of SHAPE, the probability distributions for time allocations are based partly on empirical data and partly on subjective assessments. The validity of the resulting exposure estimates are only as good as the validity of the subjective assumptions.

(2) If sufficient empirical data on MET concentrations and time allocations are available, the empirical approach based on the convolution method to be described later in this section should be preferred.

(3) It might be easier to modify the simulation models to allow for independence between MET concentrations and time-allocations than to modify the convolution method. Parametric joint distributions can be specified for C and T (allowing for possible dependence); the joint distribution (including the dependence) can be estimated from observed

data, then one can initiate the simulation using the estimated joint distribution.

An alternative approach is empirical and is called the convolution method. With the assumption that C and T are independent, we can estimate the exposure distribution by considering all possible pairs of sampling units in the monitoring sample and the time-allocation sample. From each such pairing, we estimate the exposure using the time-weighted summation formula

$$E_{im} = \sum C_{mk} T_{ik} \quad (6)$$

where E_{im} is the exposure combining the i th unit in the time allocation sample with the m th unit in the monitoring sample. C_{mk} and T_{ik} are the MET concentration and time allocation of the same units in the k th MET. We then estimate the distribution of exposures by the empirical distribution of E_{im} 's for all possible pairings of i and m . For each observation unit in the monitoring sample, say the m th unit, the outputs include a point estimate of exposure and a distribution of exposures from the empirical distribution of E_{im} 's for the fixed m and all possible i 's. Similarly, for each observation unit in the time-allocation sample, a distribution of exposures is obtained.

2.6 HUMAN GASTRO-INTESTINAL AND SKIN EXPOSURE

Using the definition of exposure detailed above, which can be paraphrased as the concentration of a substance in the vicinity of an organism, it follows that directly ingested material represents a dose of the substance rather than an exposure. The assessment of exposure consists of the measurement of concentrations of the substance concerned in samples of food and water. Problems in getting representative samples and in combining data from different food and water samples to get dose estimates are detailed in papers in the second part of this volume.

Exposure to air pollutants via skin absorption can be treated in a similar fashion to the treatment of inhalation detailed above. Also exposure via skin absorption to pollutants in water may be treated in a similar way, but such studies do not seem to have been attempted to date.

In indirect dietary exposure studies, human subjects have been sampled and their food consumption surveyed. The survey methodology described earlier in the direct approach can be applied in this area, with monitoring instruments replaced by food consumption questionnaires. Based on the measured food consumption described for the sample, the comparable food basket estimates are obtained for each individual in the sample. Individual foods or composites of them can be prepared according to a prescribed procedure, and the total diet analysed to determine the chemical. There has

been a tendency to segregate the total diet into smaller composites for analysis. Between the two approaches, the second (total diet) approach is preferable, because it captures the transformation and losses of the chemical during preparation.

2.6.1 MULTIMEDIA EXPOSURE

The very important problem of the relative importance of different exposure routes in determining the overall dose of the chemical is one that has received little detailed study in the past. One of the few studies in this area, the TEAM study conducted in the USA (Wallace *et al.*, 1987) produced results that call into question some of the current assumptions made in pollution control strategies. A study of multimedia exposure to metals and chlorinated hydrocarbons including biological indicators of exposure is being conducted simultaneously in Japan, Sweden, USA, and Yugoslavia.

2.7 NON-HUMAN EXPOSURE

It is usually impossible to directly assess exposure of non-human biota; thus some form of indirect analysis is usually required. In the past, this process has often amounted to little more than a measurement of the concentration of a chemical of interest in the environmental medium thought to be of major importance, e.g., analysis of water for a dose to a fish or of air for a dose to terrestrial plants. Some more complex studies have been conducted to assess the relative importance of alternative exposure routes, for instance, aimed at delineating the relative contribution of food in determining direct exposure of fish to organic chemicals in water, including both modeling and practical studies. In the terrestrial environment, studies have been conducted on the pathways by which radioactive elements and toxic metals move from contaminated land through food chains.

The conceptual framework of exposure and the application of statistical techniques developed for human exposure to pollutants (as summarised above) can be applied to non-human exposure. Generally, what is of concern is the exposure of a population of a given species, or of a whole ecosystem, and not that of an individual. Conceptual development in this area is needed to accommodate this and other possible differences in approach. Boundaries can be designated, for instance, for plants with leaves, roots, stem and bark, and for fungi with extracellular enzymes.

A second area that generally presents a considerable problem consists of current restrictions imposed on the specification of the physico-chemical analytical methods. These restrictions often result in a generalised operational definition of physico-chemical form such as size-separation using filtration. Operational definitions are probably of doubtful or no biological

relevance and, therefore, of little value in converting exposure data to doses. Frequently, to circumvent these and other problems (such as fluctuating environmental concentrations), residues of a chemical of interest are measured directly in the primary organism or in a surrogate. This relationship between the concentration of chemical in the organism and that in the surrounding medium is sometimes expressed as a factor independent of concentration over a given range.

In terrestrial ecosystems, plants are at times more sensitive than humans or animals to gaseous air pollutants such as SO_2 or ozone. Exposure is generally estimated by the time-integrated measurement of concentration in air at plant height. However, effects of air pollution on trees associated with the forest decline syndrome are not necessarily related to the concentration of some of the gaseous pollutants and, in general, concentrations in air are of little use in estimating exposure to "acid deposition". Significant variables are more likely to be the rates of wet and dry deposition, the latter being particularly difficult to assess.

Animals in terrestrial ecosystems are generally considered to be exposed to chemicals mostly via the food chain. Thus, the significant variable is the concentration in the plant consumed by the eater.

Soil pollution may result in uptake of undesirable chemicals by plants and occasionally by animals. The fraction of any given chemical actually available for uptake by plant roots must be known for dose/effect studies. Plant uptake is sometimes estimated by using soil/plant transfer coefficients, estimated from pot or field experiments.

2.8 USES OF EXISTING DATA

At first glance, it would seem possible to employ existing data collected from ambient monitoring networks to calculate exposures. Unfortunately, efforts to relate such data to actual exposures have been disappointing. In the air pollution field, for example, a large body of literature now exists showing poor correlations between observations taken at fixed ambient monitoring stations and the concentrations to which people actually are exposed, as measured by personal monitoring (Brice and Roesler, 1966; Ott and Eliassen, 1973; Ott and Mage, 1979; Cortese and Spengler, 1976; Wallace, 1979; Flachsbart and Ott, 1984; Ziskind *et al.*, 1982; Repace and Lowrey, 1980; Mage, 1985).

Two literature series have been published summarising some of these findings (Ott, 1982; Spengler and Soczek, 1984), and recent large scale total human exposure field studies (described below) have provided considerable information about the disparity between measurements taken at fixed ambient air monitoring stations and actual human exposure.

In the winter of 1982-83, over 1600 CO exposure profiles were obtained

in Denver, Colorado, and Washington, DC, using personal monitors and a weighted probability sampling technique of the population in each city (Whitemore *et al.*, 1984; Hartwell *et al.*, 1984; Akland *et al.*, 1985; Croce *et al.*, 1985). These studies showed generally poor relationships between the fixed air monitoring stations and actual exposures of people, particularly those persons receiving the highest exposures. The correlations between CO at fixed stations and CO measured at the same time in fixed locations were poor. For example, linear regression between composite fixed air monitoring stations and CO concentrations observed while people were engaged in various activities gave poor associations: e.g., walking on a sidewalk ($R^2 = 0.23$); driving a truck ($R^2 = 0.11$); driving a car ($R^2 = 0.04$) (Akland *et al.*, 1985). The correlations between fixed stations and personal exposures improved for locations not near motor vehicles: e.g., in suburban homes without gas stoves and in indoor recreational facilities. The most important determinants of the variation in exposure from person to person were the locations visited throughout the day (e.g. stores, offices, restaurants, garages, and occupational settings).

Visits to some of these micro-environments brought people into close proximity with sources of CO (e.g., gas stoves in residences, on sidewalks, and automobiles in garages). The main reason for the failure of fixed air monitoring stations to predict exposures accurately, therefore, is the failure to include indoor and in-transit components of exposure. Existing atmospheric dispersion models have the same fault: they model only outdoor levels and ignore the indoor or in-transit components. An important departure from modeling approaches of the past is the development of models of human exposure, such as the Simulation of Human Air Pollution Exposure (SHAPE) model (Ott, 1984), which includes human activity patterns and both indoor and in-transit components.

Even more striking disparities have been found between indoor and outdoor levels of toxic air pollutants, including many carcinogens. In the TEAM study of personal exposures to volatile organic compounds (VOC) in Bayonne and Elizabeth, NJ, pump-driven Tenax TM personal monitors were used to collect exposure data on a representative probability sample of people from 355 households. Measurements also were made of VOC in their breath. On the average, indoor levels of VOC's were 2 to 5 times higher than outdoor levels with occasional levels 70 times higher (Pellizzari *et al.*, 1984; Wallace *et al.*, 1987). The list of pollutants found indoors included benzene, carbon tetrachloride, chloroform, styrene, tetrachloroethylene, 1,1,1-trichloroethanol, and trichloroethylene. The sources are believed to be indoor furnishings, paints, solvents, hobby activities, carpets, furniture polish, etc. With such high indoor readings, it is obvious that outdoor monitors can provide only a poor indication of actual exposures, since people spend 90% of their time indoors (Ott, 1982). This has also been found for exposure estimates of inhalable particles (Repace and Lowrey,

1980). Similar discrepancies have also been reported between concentrations measured in the effluent of water supply treatment plants and water at the tap which is actually consumed by people.

Outdoor monitoring station data can, however, be relevant for exposure of plants, buildings, monuments, etc. They can also reveal areas of high exposure to an outdoor generated pollutant, and serve as an indication that in these areas external assessment would be appropriate.

2.9 RECOMMENDATIONS

More detailed recommendations to support the major conclusions presented in Chapter 1 are presented below in the same order as the earlier topics of the chapter:

(1) Probability sampling techniques and other survey research methods should be used whenever possible in designing human exposure field studies.

(2) Methodological investigations should find ways to solve the low response rate problem to eliminate it as a potential source of bias.

(3) Methodological investigations should improve our understanding of human behaviour so that a number of other factors which can lead to bias are eliminated.

(4) A program should be established to provide knowledge and technical assistance to countries that want to use the direct approach in conducting exposure assessments. This program which should be coordinated through an international organisation, should enable the development of comparable high-quality, international exposure data bases.

(5) When a continuous personal monitor is available, we recommend the enhanced personal monitoring approach because of its efficient use of monitoring data. If there is sufficient evidence that the MET concentrations in an earlier study are representative of the new population, it would be preferable to conduct a limited personal monitoring study in the new population to validate this latter assumption.

(6) If a continuous personal monitor is not available but an integrated personal monitor is available, we recommend direct personal monitoring. When neither type of personal monitors is available, we recommend using MEM as a stop-gap measure. It should be recognised that until a satisfactory solution to the problem of sampling micro-environments is available, the MEM can at best be viewed as a proxy measure of actual exposures since it is difficult to assess the representativeness of the estimated exposures. If sufficient empirical data for concentrations and time-allocations are available, the convolution method is recommended provided the independence assumption is found to be valid. For gaps in empirical data, it may be necessary to use subjective assessments and appropriate simulation models. The results of the estimated exposures from the simulation models need to

be interpreted carefully. In particular, it is important to conduct sensitivity analyses to assess the impact of perturbations in the subjective input to the models.

(7) Obvious approaches to improve our ability to assess human exposure are the development of better measurement instruments and of biological markers of exposure, both of which will be discussed in detail in this volume. Based on recommendations for EPM, continuous personal monitors are preferable to integrated personal monitors. Another step is the development of generally applicable time-allocation data bases. Many pollutants share similar sources; for example, CO and NO₂ are both generated mainly from combustion. Therefore, a time-allocation data base for a CO study that is carefully designed can be revised for a future NO₂ study, assuming that the time-allocation of the former population is representative of that in the latter population. The development of a multipurpose time-allocation data base to serve the needs for several species of pollutants can be a cost effective approach.

(8) Reliance of the indirect approach on the assumption that the MET concentrations C and the MET time-allocations T are independent is restrictive. In any application of the indirect approach, it is crucial that the assumption be validated empirically. Furthermore, it is important to develop more general analytical methods which allow for dependence between C and T .

(9) The sampling of micro-environments in MEM is another important general methodological research need for the indirect approach. Two preliminary proposals using weighted sampling schemes and simulated human activities need to be implemented and validated in field studies (Duan, 1985). Additional approaches are encouraged.

(10) Previous multimedia field studies of volatile organic compounds have revealed surprisingly high personal and indoor exposure; additional field studies should be conducted to identify the sources and factors responsible for these exposures. These data and methodologies should be reviewed by various countries for applicability to their populations. Finally, similar field studies should be conducted of VOC exposures in other cities to determine the geographical variability of these findings and the influence of lifestyles, housing characteristics, and occupational factors on VOC exposure. Suitable measurement methodologies also exist for inhaled particles, but none has been deployed in large-scale population exposure studies. Existing monitors for inhalable particles should be deployed in large-scale population exposure field studies using the techniques mentioned earlier. The chemical species should be analysed in this field study, and concurrent measurements of these compounds through food, drinking water, and other relevant routes should be undertaken.

(11) Multimedia exposure studies also are needed of pesticides and polynuclear aromatic hydrocarbons (PAH or PNA).

(12) Data on human activity patterns (i.e., "time budgets") are important for constructing human exposure models since they provide critical input to the models. Existing activity-pattern data bases usually were collected for purposes other than exposure estimation and seldom provide the information needed; thus, field studies are required to collect relevant data on human activity patterns—i.e., the micro-environments people visit and the activities they perform over a 24-hour period using diary approaches.

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