

Exposure Estimation

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ABSTRACT

The estimation of exposures of ecosystems and man to environmental pollutants requires wide knowledge of the characteristics of sources, substances, environment and receptors. The general procedures for exposure estimation are presented, including the concept of exposure, environmental transport and behaviour, and metabolic behaviour.

Several models are used to describe dispersion of pollutants in the environment and their behaviour within ecosystems and within the human body. A sequence of modelling operations may be employed, for example, air dispersion or aquatic transport models to describe initial movement of the pollutant from the source followed by food-chain or ecosystem models to determine further transfer to the receptor.

The results of modelling are used in conjunction with the results of monitoring. Monitoring programmes provide basic data for input to and validation of models. The general features of environmental and biological monitoring are briefly discussed.

Exposure estimation by means of modelling and monitoring make up the first stage of pollutant assessment. It is necessary to combine these results with information on toxicity and effects of exposure to pollutants. The complete exposure and effects evaluations provide hazard assessment of the pollutant and point to the measures required for prevention or control of damage to human health and to ecosystems.

1 INTRODUCTION

The hazards of chemicals released by man into the environment depend most generally on the source production and release rates, the environmental dispersion and persistence, bioaccumulation and toxicity. In this paper the methods for estimating exposure of ecosystems and man are discussed, including environmental and biological monitoring.

Evaluation of exposure of biological organisms, including man, to chemicals in the environment requires understanding of the environmental behaviour of

different substances, their movements, interactions and transformations in various media. Although the basic cycling of trace substances in the environment is reasonably well understood, the physical, chemical, biological and interaction properties of specific chemicals cover such a wide range that each substance in each environmental situation requires a unique assessment. Due to inherent variabilities and incomplete data bases, particularly for new chemicals, uncertainties in the exposure evaluations cannot be avoided.

Information required to describe environmental behaviour of chemicals includes physical and chemical properties of the substance and characteristics which determine environmental movement and fate. These properties include solubility, reactivity, volatility, degradation potential, bioavailability as well as some attributes of dispersive media such as wind speed, flow rate, pH, presence of organic matter, particulate matter and biological activity. A complete specification of properties is necessary for detailed mathematical modelling, but even an incomplete listing may indicate similarities to the behaviour of better known substances.

A great variety of models have been formulated to describe the temporal and spatial variations of concentrations of chemical substances as they move through the environment. These models generally reflect cycling of substances in the global, regional or local environment, or within ecosystems, or they may describe the mechanisms or results of interactions of environmental components. Models are simplified representations of physical and biological systems. Simple models are an aid to both understanding the interplay of system components and describing generalized behaviour. More advanced models, which can be validated with experimental or field data, can gradually narrow the degree of uncertainty in specifying interactions and in predicting future changes.

In addition to knowledge of environmental behaviour of chemicals, information on the activity and consumption habits of the organism or individual is required in exposure evaluations to determine the intake amounts. Metabolism affects the distribution and retention of chemicals and their byproducts in sensitive tissues or organs where effects may ultimately occur. Information on source release patterns is required to estimate the levels of chemicals in the environment and to relate to monitoring and control programmes.

2 THE CONCEPT OF EXPOSURE

Exposure is the quantity against which the end effect in any biological system under consideration is tested. Only with exposure to an agent can there be an effect occurring above a spontaneous level. With increasing exposure the effect appears to follow some kind of functional 'exposure-response' relationship. In practice, more specific definitions of the exposure quantity are required.

'Exposure', and the closely related and often interchangeable word 'dose', are variously used terms in toxicology and environmental science. Classically,

'exposure' may simply refer to the presentation of a substance or physical agent to a receptor and 'dose' to the amount administered or taken in via inhalation, ingestion, injection or absorption through the skin. 'Dose' has been further specified as the amount reaching the site of effect (Nordberg, 1976), but 'exposure' is also used in this context (Butler, 1978). The terms intake and uptake overlap these usages, intake referring to the entry amount and uptake the absorbed amount.

In radiation studies 'exposure' has a restricted definition, namely the electrical charge produced by ionizing radiation per unit mass of air. The 'absorbed dose' is the energy absorbed per unit mass of the irradiated material. Absorbed dose is the more widely applied term and 'dose-response' relationships are established to relate functionally the energy absorbed by the irradiated object with the response observed.

In the case of chemical compounds, a useful definition of exposure is difficult because of the varied nature of interactions and uncertain mechanisms which produce effects. It has been proposed to define exposure as the number of primary chemical events leading to the final effect (Ehrenberg *et al.*, 1974). It is, however, only in rare cases that the nature of such primary events is known and their quantification possible.

It has also been suggested that the exposure to chemicals be expressed as radiation equivalence by equating the end effects. This has only an empirical basis and does not appear to be entirely satisfactory. This concept and the various limitations to its application have recently been reviewed (Moustacchi and Latarjet, 1979).

Exposure to chemicals is best defined in terms of concentrations and the duration of the substance's presence at the point of interest. Concentration is the parameter most frequently available, either in the external environment or internally following intake and absorption. To give exposure some biological meaning, the concentration should, if possible, be expressed at the level of the target biological structure.

In addition to concentration, it must be recognized that duration of exposure is an important factor in the manifestation of effects. This reflects either the time for accumulation of the substance or of metabolites to sufficient levels to induce damage or the accumulation of damaged elements that eventually inhibit proper functioning.

The most direct combination of concentration and duration in a definition of exposure is the time integral of the concentration.

$$E = \int_0^T c(t) dt$$

The concentration may be in an environmental medium, such as air or water, or it may be in the organism or at a specific point in the biological system. This

formulation of exposure is particularly useful in describing the movement of chemical substances in the environment. The transfer between environmental reservoirs is described by linear, first-order kinetics. The integrated concentrations and also integrated fluxes maintain constant relationships in this case.

In terms of effects, this definition of exposure must be used with some caution. The various combinations of concentration and time giving equal exposure, for example high concentrations of short duration and low concentrations of long duration, may not be associated with equal occurrence of adverse effects. This is the case if threshold levels are known to apply. On the other hand, there are many chemicals which are found to be harmful even at the lowest levels. It may then be reasonable and conservative to assume a linear relationship between exposure and effect.

Other functional forms of exposure relationship may be required in the evaluation of specific effects in particular biological systems. Some of these forms, involving exponential or power functions of time, have been discussed by Piotrowski and Buchanan (1982). It will be necessary for toxicologists to give further consideration to the concentration-time relationship, i.e. the expression of exposure, which is most closely associated with the occurrence of effects.

3 ENVIRONMENTAL TRANSPORT AND BEHAVIOUR

Environmental behaviour of chemicals is most often described by representative models. Modelling begins with the specification of the compartmental arrangement which reflects the system components. For first stage modelling of the regional or global environment, very broad compartment identification may be sufficient: atmosphere, land, freshwaters, ocean. At a later stage, more sensitive areas may be considered, for example, estuaries or specific ecosystems. Simplified models have advantages in utility. Increased complexity is justified only for substantial improvements in reliability or resolution of the results.

Models can be broadly classified as either time-dependent or time-independent in their basic formulation. Time-dependent models give the dynamic behaviour of the chemical substance in the environment. First-order kinetics is generally assumed and a system of differential equations describes the changes in compartment contents. Solutions may be obtained directly, if possible, or simulation of the transfers occurring may be carried out in small time sequences. This may be referred to as a systems analysis method. It is important that the compartmental arrangement be correctly formulated and the various transfers and interchanges be accurately specified to reflect the proper environmental behaviour of the chemical. A large data base is required to evaluate this type of model, but the evaluation is most complete with amounts and transfer rates given for all points and at all times.

A time-independent description of the behaviour of a chemical can be made using a more limited data base. In this case, the objectives are to determine the

partitioning of the amounts of chemicals in pathway movements and to estimate the amounts which ultimately reach the receptor. The relationships between compartmental amounts are expressed as the ratios at steady state, called the concentration factors. The time-independent approach is often referred to as the concentration factor method. A wider application involving cumulative transfer per unit input is called the exposure commitment method.

Examples of the two basic approaches to modelling are given by Roberts *et al.* (1981), with a simplified kinetic model of the aquatic environment, and by Bennett (1981) using the time-independent exposure commitment method to evaluate terrestrial and aquatic pathways of transfer. Such examples may be the basis for generalized, systematic examination of the fate of chemicals in the environment. Particularly for screening large numbers of chemicals, it is necessary to determine broad aspects of behaviour, such as the likely sequestering compartments and hence the likely receptors of higher exposures, the persistence pattern of chemicals in each compartment, and the main exposure pathways. It is then necessary to consider other aspects of the chemistry, toxicology and ecology of the chemical and the environment—the potential for degradation, the toxicity of primary and degradation products, interaction factors, tolerance of species to exposures, secondary receptors, etc.

A sequence of modelling operations may be conducted to complete an exposure assessment for a chemical. For example, air dispersion or aquatic transport models may be applied to the initial movement of the chemical from the source to the point of interest. Food-chain models may then be used to determine the further transfer of the chemical to the receptors. This general approach has been particularly developed and applied to assess the environmental behaviour of radionuclides in the environment (UNSCEAR, 1977; CEC, 1979). The methods are generally applicable. It is not possible here to make a comprehensive list of models, but a brief discussion of the main types may be useful.

3.1 Air Dispersion Models

Atmospheric transport models reflect the impact of meteorological factors on concentration of trace substances in air (Turner, 1979). Inputs to the models include meteorological data and emission estimates. Most commonly, air dispersion models are based on the Gaussian solution of the atmospheric diffusion equation. Utility of the model was enhanced by the Pasquill-Gifford methods of selecting parameter values (Gifford, 1961; Pasquill, 1961). Dispersion estimates can be made for a wide variety of meteorological conditions, i.e. wind speed and direction and atmospheric stability. Estimates for particular averaging periods are obtained by weighting the results according to frequency of occurrence of particular conditions (Turner, 1970).

The Gaussian plume model accounts for downwind transport and dispersal of

substances due to normal atmospheric turbulence. A particular result depends on constant wind and stability conditions and, therefore, there are limits on time and distance validity. Generally, predictions are considered reliable only within a few tens of kilometres from the source.

Estimates of the dispersion of substances to greater distances may be made by analysis of the trajectories of the air masses which pass over the release point. An example is the trajectory model of Heffter and Taylor (1975). Trajectory models are referred to as Lagrangian or moving cell methods. An alternative approach is the fixed coordinate method, referred to as Eulerian or grid models. These provide a numerical solution to the three-dimensional advection-diffusion equation. The particle-in-cell technique providing multiple trajectory estimates may be used in conjunction with grid models (Lange, 1978). These models can become quite complex requiring large computer capacity and must be backed by detailed wind data. However, account can be taken of a wide variety of changing conditions and of complex terrain.

Modelling the dispersion of reactive gases, such as nitrogen oxides and hydrocarbons, requires information on the chemical kinetics as well as the air flow behaviour. Interacting aerosol-gas systems, notably the SO_2 -sulphate-sulphuric acid conversions, are complex and difficult to represent in atmospheric transport models. Photochemical modelling uses trajectory and grid techniques (Turner, 1979).

It is necessary to account for various removal processes of chemicals from the plumes and for chemical transformations which may occur during transport. This is done by estimating effective source strengths or estimating depletion at various stages of the transport. Wet deposition refers to removal of substances by precipitation falling through the plume (washout) and to attachment in droplets in the rain forming layers (rainout). Dry deposition occurs by impaction of aerosols on the underlying surface. The deposition velocity, which is the ratio of aerosol deposition per unit time per unit area to the average concentration of the aerosol in air, has been widely used (McMahon and Denison, 1979; Schmel, 1980).

3.2 Aquatic Transport Models

Aquatic transport of chemical substances is governed by general water movements and sedimentation processes. Models are categorized according to whether releases are to rivers, lakes, estuaries or coastal seas. It may be assumed that relatively rapid mixing occurs in the receiving water volume or that equilibrium is achieved. Otherwise flow characteristics can be modelled if hydrological information is available.

River models are based on a one-dimensional equation accounting for diffusion (mixing) and advection (movement with water mass), accounting for sources and sinks including sediment interactions. Alternatively, the river may be

considered to be a series of discrete compartments, or an empirical function may be found relating the concentration of the substance with downstream distance from the sources (CEC, 1979).

Lake models employ similar hydrographic and sedimentation data. It is usually possible to define the turnover time of water in the lake and the effective residence time of the dissolved chemical. Deeper lakes may require stratified models with seasonal mixing of the layers.

Estuaries have a number of unique features, notably the change in salinity from the river to the sea and the tidal influences. Estuaries are sediment-rich areas. Compartment modelling may be possible for water sections, balancing input and outflow from specific areas.

The simplest models for coastal seas are again compartment models, where a volume of assumed uniform mixing is defined. The sedimentation and tidal influences are variable and the local and large-scale water movements are site-specific.

The degree of interaction of chemical substances with sediments is indicated by the value of the sediment-water distribution coefficient, K_d , defined as the ratio of the concentration of the chemical in the sediments to that in the water. Chemicals are adsorbed to suspended sediments and to bed sediments. There are many factors which affect the rate of attachment to, or release from, sediments such as the pH of the water and salinity.

An important input of chemicals to aquatic systems is surface run-off from soils to lakes and streams and downward penetration through soils to ground water. Downward migration of chemicals in soils is due to leaching with rainfall or irrigation water input. Factors affecting the movement include solubility, soil properties including potential for adsorption to soil minerals or organic material, and water influx. Chemicals soluble in water and poorly adsorbed by soil components are the most mobile. Downward movement in water occurs in preference to surface run-off until the surface layer is saturated.

Migration of chemicals through the soil column can be represented by transfers between compartments of varying depth (CEC, 1979). Methods of modelling the sorption processes occurring in soils have been reviewed by Travis and Etnier (1981). Models of movement of chemicals in run-off require consideration of the properties of the chemical and soil, the hydrology and soil loss. Models describing pesticide dynamics in soils have been discussed by Crawford and Donigian (1973) and Rao and Jessup (1982).

3.3 Food-chain and Ecosystem Models

Transfers along terrestrial and aquatic food chains are represented as compartmental sequences with transfer factors or concentration ratios expressing the relationships at steady state. These have been particularly developed and applied to radionuclide environmental behaviour (UNSCEAR, 1977; CEC, 1979), for

example, soil-pasture grass-cow-milk-man. The details of the model depend on the availability of data. The aquatic models often proceed in one step from water to aquatic organism with use of the concentration factor. If the concentration factors have not been measured, there are suggestions for estimating them from chemical properties (solubility, octanol-water partition coefficient) and the physiology of the organism (Neely, 1979).

Linear system models may also be formulated to describe food-chain transfers. The interlinking transfer rate constants must be evaluated. The solution then gives the concentration variations as a function of time (Booth and Kaye, 1971).

Ecosystem theory, involving examination of all the aspects of stress, stability, energy fluxes, nutrient cycles, carbon metabolism and trophic structure, is being applied to assessment of chemical pollutant behaviour. The steps in model formulation and implementation have been discussed by several authors (e.g. Goodall, 1972, 1974; Reichle *et al.*, 1973; Wood, 1974; Swartzman, 1979). Information availability is the major limiting factor in direct applications of large-scale ecosystem models.

3.4 Environmental Monitoring

Exposure of organisms and man to chemicals in the environment can be evaluated with most certainty by direct measurement of concentrations and fluxes, provided the sensitivity of methods is adequate. Rather extensive and comprehensive measurements would be desirable, but limited resources mean that selective measurements must be made to provide essential information. Monitoring and modelling programmes are mutually supportive. Modelling studies suggest the more important types of measurement and extend the measurement results. Monitoring programmes provide basic data for input to and validation of modelling efforts.

Essential features of monitoring programmes include site selection, sampling methods, basic measurements to be undertaken, frequency of measurement and interpreting procedures. Either index or random sampling may be considered appropriate (NAS, 1975). In index sampling, judgments of representativeness and availability predominate in the selection of materials. It is assumed that index materials may reflect the levels in the broader population of interest and the interrelated levels in associated compartments. An example is the selection of a single species of fish to represent all fish or perhaps all aquatic biota. Random sampling is generally appropriate only in more limited monitoring programmes. Examples are analysis of various foods to determine dietary intake of particular substances and area sampling of soils to determine average concentrations (NAS, 1975). Usually, some form of stratified sampling is undertaken, which gives more attention to the more important items or regions to be investigated.

The measurements in monitoring programmes must be appropriate to the

purposes, reflect spatial and temporal variations, provide details of physical and chemical forms and be reported with relevant parameters for other media properties. An integrated approach to monitoring is required—associated multi-media and multi-substance analyses—to evaluate the behaviour of chemicals and their interactions in the environment.

Monitoring data should indicate the changes with time of concentrations and media properties to establish whether equilibrium conditions have been reached or to reflect trends in accumulation or diminution of the amounts of chemicals or degradation products. It is important that not only immediate exposures be determined, but the build-up of chemical substances in soil, sediments or other sinks be monitored and long-term potential transfer to organisms and man be evaluated.

4 METABOLIC BEHAVIOUR

4.1 Metabolic Models

Metabolic models are used to describe the relationships between intake of chemical substances and their absorption, distribution, retention, transformation and excretion by biological organisms. The models may be useful in relating intakes to measured concentrations in body fluids, such as blood and urine. However, their primary use is to estimate the exposures to critical organs, that is, those which accumulate the highest concentrations or are the most sensitive to damage or dysfunction.

As in general modelling, time-independent or time-dependent metabolic models may be used. At steady state, the concentration factors give simple expression of the disposition of intake amounts. These are ratios of the concentrations of chemical substances in the organism and in the intake amounts. It may be unrealistic to assume that the measurements are made at steady state, particularly if there are long-term transfers or wide variabilities. The results apply to the particular environmental conditions, but wider applicability can usually be assumed for reasonably comparable circumstances.

Compartment models of more or less detail and complexity are used to describe the metabolic processing of chemicals. The compartments may represent the physical structure of the organism, such as primary organs and tissues, or they may represent the various components of retention. Retention equations consisting of a series of exponential terms are very useful in reflecting overall metabolism. The parameters are determined from excretion measurements or from analysis of tissue content at various times after an amount has been administered. The half-times of retention and the effective mean residence times of the chemical in the organism can be determined.

Models have been developed which reflect physiological processes following intake. These include a lung model and a model for the gastrointestinal tract. The

lung model, formulated by the Task Group on Lung Dynamics (ICRP, 1979), divides the respiratory system into nasopharyngeal, tracheobronchial and pulmonary compartments. Deposition of inhaled particles in the compartments depends on particle size. Clearance from the compartments is by mucociliary transfer to the gastrointestinal tract or absorption in blood. The model gives the pathways and clearance rates for several classes of particle solubilities. The classes of compounds are designated D, W, and Y for clearance times from the pulmonary region of the order of days, weeks or years. For the amounts absorbed into blood, further assumptions can be made regarding the distribution of the chemical substance to organs or to excretion. The ICRP lung model is very widely used.

The behaviour of ingested materials can be described by a physiological model such as the one by Eve (1966), which was developed for purposes of radiation dosimetry. The residence time of ingested material in each section of the gastrointestinal tract is specified. For radionuclides, radiation doses are received during these periods. For chemical substances, the passage times are less important than the fractional amount absorbed into extracellular fluids. Summaries of absorption values and other metabolic data for chemical elements are given by ICRP (1979).

Multicompartment models have been formulated to represent the metabolism of specific chemical substances, for example an eight compartment metabolic model for cadmium (Kjellström and Nordberg, 1978) or a five compartment model for lead (Bernard, 1977). Reviews of this and other aspects of the modelling procedures are given by Gehring *et al.* (1976), WHO (1978), and Camner *et al.* (1979).

4.2 Biological Monitoring

Biological monitoring is the examination of organisms to estimate environmental levels and/or effects of pollutants. Organisms accumulate or respond to environmental substances and are frequently sensitive indicators of the presence or effects of these substances.

The biological samples include plants and animals from the region of interest and from background areas. Most often assessed are the species diversity, abundance, productivity and biomass. Functional variables, such as carbon fixation, may also be monitored. In some cases, attention is focused on key or critical species with the assumption that associated species will be affected in proportion. Community structural analyses involve counts of species present and individuals per species. Because of the interdependent system of species within ecosystems, effects on one part may ultimately affect the whole.

Often implied in biological monitoring is the sampling and analysis of body fluids and tissues of man. Blood, urine, hair and faeces are most easily obtained. These results give indirect indications of accumulation of substances in the body

and point to altered functioning or other effects. Tissue samples may also be obtained, primarily at autopsy, for direct analysis of chemical content or for evidence of damage.

The results of biological monitoring may be used with the metabolic models which describe the processing of materials by organisms. These include absorption factors for specific intake modes and forms and the distribution and retention kinetics of the chemicals in the body. Either dynamic models or steady-state concentration factor measurements may be available.

Special precautions must be taken in the sampling and analysis of biological materials. The levels of chemical substances to be investigated may be quite low, and care must be taken to avoid contamination from sampling equipment.

The difficulties in obtaining accurate analytical results for biological materials have stimulated an interest in collaborative studies. Interlaboratory comparisons have been undertaken, but the results of these are often quite disappointing, showing wide differences even for quite common measurements, for example lead, cadmium and mercury in blood, urine and water in one particular intercomparison (Lauwerys *et al.*, 1975). The need for strict analytical quality control has been frequently pointed out (see for example WHO, 1979).

An international programme of biological monitoring has been initiated by WHO and UNEP to assess human exposures to selected toxic pollutants. In this project the levels of lead and cadmium in blood, cadmium in kidney cortex and certain organochlorine compounds in breast milk are being determined in selected population segments in 10–12 countries in different parts of the world (Slorach, 1982; Friberg and Vahter, 1983).

Results from biological monitoring programmes may be used directly in assessing exposures of receptors to chemical substances. These measurements, however, do not provide associations with the individual sources of exposures, and therefore results from such studies must be correlated with the results of environmental monitoring programmes and evaluations of the receptor locations and activity patterns. Further considerations of biological monitoring of metals and other compounds have been given by NAS (1975), Berlin *et al.* (1978) and Cairns (1979).

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