

Assessment of Human Exposure to Chemicals from Superfund Sites

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Introduction

Assessing human exposure to chemicals from Superfund sites requires understanding and applying knowledge from a number of scientific disciplines and gathering large amounts of data. It is necessary to know how chemicals behave once they have been deposited in these sites, how they behave once they are released from the sites, and how they behave once they come into contact with humans. This complex array of information must be gathered for a large variety of different chemicals and for human populations that vary significantly in important characteristics. Considering the importance of local conditions, assessments for each Superfund site must be location specific and so must depend on emission, transport, and exposure data gathered at that particular site using the population at greatest risk.

To characterize this complex situation, generic models are being developed that approximate the physicochemical behavior of chemicals and describe their environmental distribution and fate. In addition, more information is being gathered about behaviors of individuals to better estimate human exposures. At the same time, research is underway on improved methods for gathering the data necessary both to test the models and to provide the requisite information for evaluating potential exposures and health risks from specific sites. The focus of this report on the Conference on Assessment of Human Exposure to Chemicals from Superfund Sites, held at Michigan State University on June 5-6, 1990, is current progress with such model development and with

data collection methodology. The strengths and limitations of current knowledge and theories are illustrated with case studies provided by the participants in the meeting. At the end of this summary report, the reader should have an appreciation of what is known, what is not known and what needs to be done to bridge the gap between the two. A summary of the meeting program is provided in the Appendix.

Environmental Distribution

The environmental distribution of chemicals from Superfund sites depends on a number of intermedia transport processes that were described by Cohen (1). Three such processes by which substances may leave the site and move to the different environmental compartments are runoff to surface water, leaching to groundwater, and volatilization and respiration to air. In addition, resuspension of contaminated dust to the air may be important at some sites. Redistribution from these initial compartments can occur over time, and this can be a global process for persistent chemicals. Describing and predicting the movement of these chemicals require knowledge of the factors that influence migration between soil and air, soil and surface water, soil and groundwater, surface water and air, and sediment and surface water. In addition, partitioning between each compartment and biota can contribute significantly to total human exposure. In particular, bioaccumulation and biomagnification of lipid-soluble chemicals can lead to high contaminant levels in organisms that are part of the human food supply. The following sections address current knowledge and ongoing research on transport processes between environmental compartments.

Soil to Air

The exchange of pollutants between the soil and the atmosphere is determined largely by volatilization (2-5). Recently, Cohen et al. (6) and Cohen and Ryan (7) demonstrated that contaminant transport toward the surface is significantly affected by moisture and temperature variations as a result of their impacts on diffusion, convection-dispersion and partition coefficients. The transport of pollutants to the atmosphere may also occur by the resuspension of particulate matter (8).

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Soil to Groundwater

Although many types of chemicals are found at Superfund sites, organic chemicals are often of greatest concern, particularly if they leach to groundwater below the site. T. Voice (Michigan State University) discussed subsurface processes affecting groundwater contamination. If these compounds are immiscible or nonaqueous-phase liquids (NAPLs), they may float on top of the water table or move down until they reach an impermeable layer, depending on the density and hydraulic gradient of the NAPL (9,10). When the NAPL contacts groundwater, soluble components can dissolve and move with the groundwater, potentially contaminating large portions of an aquifer.

As the NAPL migrates through the soil, small amounts are immobilized due to capillary forces. This residual NAPL can also pose a threat because it will come into contact with infiltrating precipitation and soluble components can be carried down to groundwater. Thus, even if the bulk of the material is sequestered in soil, those components that dissolve in water can slowly move downward and contaminate groundwater over a prolonged period of time.

Mathematical models have been developed to quantitatively describe the movement of dissolved, gaseous, and liquid organics in the subsurface environment (11,12). However, to apply these models successfully, more understanding is needed of the processes governing the movement of NAPL in both the saturated and unsaturated zones and partitioning of compounds between NAPL, air, water, and soil surfaces. Laboratory research is underway to gain this understanding, but much more work will be required to refine and validate models such that these models will provide adequate descriptions of contaminant movement into groundwater and thus potential human exposure.

Sediments/Surface Water and Biota

Metals are common contaminants at Superfund sites. D. Di Toro (Manhattan College) described some recent research on these substances. Metal wastes often have been discharged into surface water and have accumulated in sediments. It is necessary, therefore, to be able to predict bioavailability and toxicity of metals present in sediments. The situation is complex, as evidenced by experimental data indicating that equivalent metal loadings in sediments may induce very different toxicities in native biota at different sites.

It appears that bioactivity in the pore water associated with sediments governs chemical toxicity to sediment biota (13). It has recently been shown that labile sulfides have a controlling influence in this process. The acute toxicity of cadmium in sediments increases dramatically when the molar ratio of cadmium to acid-volatile sulfides concentration exceeds unity (14). This relationship also holds for nickel toxicity. Additional studies revealed that similar molar ratios produce similar toxic effects in both marine and freshwater sediments.

These laboratory findings were tested at a Superfund site, and the sediment samples taken at the site exhibited the same properties as were demonstrated in the laboratory experiments. Thus, it appears that acute biological activity of metals in sediments can be predicted from molar ratios of metals to sulfides. However, relationships between these parameters and chronic toxicity have not yet been investigated.

Although *in vitro* toxicity studies provide some information

about bioavailability, studies of the uptake of pollutants by fish provide information more directly applicable to human exposure because consumption of fish may represent a significant route of human exposure to chemicals from Superfund sites. One approach to uptake, described by R. Erickson (U.S. Environmental Protection Agency), uses physiologically based pharmacokinetic models to describe and predict contaminant levels in fish. The models consider pollutant uptake from gill surfaces and the gastrointestinal tract because these are the most significant routes of entry to the fish.

Laboratory studies of fish toxicokinetics have shown that uptake of chemicals at fish gills is affected by chemical lipophilicity, pH, fish size, dissolved organic matter, and dissolved oxygen (15). Predictions of uptake can be based on independently measured parameters such as respiration, cardiac output, fish morphology, diffusion coefficients, and chemical speciation. Absorption in the gut appears to depend on comparable parameters. Uptake estimates can then be used in conjunction with relative mass, chemical partitioning information, blood perfusion rates of various tissue compartments, and estimated rates of metabolic transformations to provide toxicokinetic models for contaminant movement and fate in whole fish (16).

Appropriate laboratory data can also be used directly in field assessments. An example is the investigation of the relationship between dibenzodioxin and dibenzofuran contamination of Lake Ontario fish and landfills on the Niagara River. 2,3,7,8-Tetrachlorodibenzo-*p*-dioxin (TCDD) uptake by lake trout was studied in laboratory chambers containing contaminated sediments and food. First-order rate constants derived from these experiments were used in conjunction with measured contaminant concentrations in lake sediments and forage fish to accurately predict observed TCDD contamination of Lake Ontario lake trout. Using this approach, it was not necessary to accurately characterize water-column TCDD concentrations (17).

Air to Other Media

Although much chemical movement is downward to groundwater or lateral to surface water, significant amounts of Superfund chemicals move upwards, volatilizing or being transported on dust into the air. H. Niki (York University, Canada) discussed the distribution and fate of these substances in the atmosphere (18-22). This movement depends on a number of factors including source parameters, meteorological conditions, and the physicochemical characteristics of the chemicals released. Both long- and short-range transport need to be considered, as illustrated by the presence of industrial waste chemicals in Arctic haze and the buildup of such substances in the food chain in Arctic regions.

The initial mixing of volatilized chemicals depends on factors such as the source configuration, terrain, gaseous and particulate composition of the air, and the nature of the chemicals. Further mixing is sensitive to the degree of contact between the gaseous and aerosol components. The size of the particulates and the vapor pressure of the chemical are critical to partitioning between the particulate and gaseous phases.

Chemicals can be scavenged from the atmosphere by either wet or dry deposition. Dry deposition occurs by diffusion, impaction, or sedimentation. Snow, ice, rain, and clouds are the vehicles of wet deposition. Neither wet nor dry deposition is well

understood although some factors, such as air/water partitioning, are known to play important roles.

Models that incorporate the above factors and processes have been developed to describe both transport and fate in the atmosphere. However, validation of the models has been difficult due to spatial heterogeneity of contaminants in the atmosphere and analytical difficulties in measuring low vapor concentrations and low concentrations of contaminants adsorbed to particulates. New approaches and techniques will be needed to overcome these difficulties and to achieve the level of understanding necessary to predict atmospheric fate and transport of chemicals from waste sites.

Environmental Transformation

Once chemicals are released from Superfund sites, they may persist unaltered or they may be transformed into other substances by chemical, physical, or biological processes. Understanding the types and rates of transformations that substances can undergo and how these vary under different environmental conditions is critical to assessing human exposures. This information is also valuable for designing remedial approaches that can take advantage of, or improve upon, these naturally occurring processes.

Chemical Transformations in Air

Niki also discussed chemical transformation of gaseous-phase chemicals in the atmosphere, a process that is governed mainly by free-radical chemistry. Photochemical transformation of ozone creates hydroxyl radicals in significant numbers, and these are the key reactive species in atmospheric reactions. Although such reactions may result in the degradation of toxic chemicals, they may also result in the conversion of less toxic compounds to more toxic ones (e.g., pyrene to nitropyrene).

Many chemicals of interest are not in the gaseous phase but instead are found adsorbed to particles in the atmosphere. Reactions occurring on particle surfaces have not been well studied. However, surface reactions involving hydroxyl radicals can be observed in laboratory studies. Further work is needed to determine if these same processes can be detected in the environment.

Microbial Transformations

Microbial action is an important natural process leading to the degradation of soil contaminants, including some of those emanating from Superfund sites (23–25). To illustrate the processes involved in such transformations, T. Vogel (University of Michigan) used the microbial breakdown of halogenated hydrocarbons as a model. There are four types of reactions potentially involved in such biological degradation processes: substitution, elimination, oxidation, and reduction. The half-lives of the halogenated hydrocarbons are dependent on the characteristics of the specific members of the group and the operative mechanisms of degradation for each. For substitution or oxidation reactions, the half-life increases with increasing halogenation. In contrast, half-lives are generally shorter for halogenated hydrocarbons that undergo elimination and reduction reactions (25).

The type of microbial degradation also depends on the presence or absence of oxygen. Under aerobic conditions,

oxygen is the preferred electron acceptor; however, under anaerobic conditions, halogenated compounds may be used as electron acceptors. Knowledge of these processes can be used in designing clean-up operations. For example, in the Hudson River sediment, the first step in degradation of polychlorinated biphenyls (PCBs) should use anaerobic organisms to reduce the degree of halogenation. This can then be followed by aerobic degradation of the lower-chlorinated products.

Many contamination sites are quite complex involving mixtures of pollutants and a large variety of microbial forms. In addition, abiotic as well as biotic processes generally occur simultaneously at a site. This makes it more difficult to evaluate the degradation processes that may occur among competing chemical and microbial species. However, prediction of degradation products is important not only for clean-up but also to assess potential exposure to these compounds, which may be more toxic than the parent chemicals deposited at the site. Thus, improved site evaluation methods and increased understanding of both biotic and abiotic degradation processes are needed.

Exposure Monitoring

In addition to using modeling of environmental distribution and fate, the types and amounts of human exposures to chemicals at Superfund sites can be assessed by direct environmental monitoring in the exposure zone of the individual by using wildlife sentinels and by analyzing human fluid and tissue samples. To use these techniques most successfully, basic knowledge about absorption, distribution, and fate of chemicals in living organisms is needed.

Physiological Pharmacokinetic Models

Physiological pharmacokinetic (PBPK) modeling, developed to describe uptake, storage, metabolism, and excretion of chemicals, was discussed by R. Dedrick (National Institutes of Health). Such models combine mathematical equations describing the dominant physiological processes and biochemical interactions into an overall expression of the dynamics of the internal dose (26–30). By means of this approach, the fate of absorbed compounds can be described over time. Data from *in vitro* and *in vivo* experimental systems can be used in combination with human physiology and anatomy to predict the movement of chemicals within the human body. Until recently, this methodology has been used primarily for drugs but now has also been applied to a limited number of environmental contaminants.

One problem in applying this approach to Superfund site exposures is that it has been validated only in situations where exposure was high and consistent. In contrast, environmental exposures are usually at low levels and vary greatly over time. This can have a significant impact on the use of the pharmacokinetic models. For example, it is not known if low-concentration nonlinearities in the kinetics of toxicant behavior must be incorporated to describe events occurring at environmentally relevant doses. Also, changes may have to be made to account for the differences between environmental routes of exposure and those used in laboratory validation studies. Indeed, quantitative comparisons and predictions among various routes of administration present some of the greatest opportunities for pharmacokinetic modeling.

Application of physiological pharmacokinetic models to human exposure assessment must consider differences between experimental animals and humans. For example, the ratios of many physiological processes to body weight decrease as size of the animal increases. A 6-hr exposure in rodents may be very different from a 6-hr exposure in humans. Also, growth rates vary from species to species, and these differences can have significant impacts. For example, PCB concentrations in fat may decrease more due to growth than to elimination in some situations. Thus, while applicable pharmacokinetic models exist or can be developed, additional work will be required to validate these models for use in evaluating human environmental exposures such as those occurring at Superfund sites.

M. Morgan (University of Washington) examined the use of models to describe pulmonary absorption of chemicals. Although absorption can occur at a variety of sites on the respiratory tract surface, the primary site is the alveolar region of the lung. A variety of physiological and physicochemical factors influence the rate of absorption (31). One of the more important is the blood-to-air partition coefficient, which reflects the solubility of the compound in blood. The higher this coefficient, the greater the absorption.

It can be shown experimentally that this coefficient is independent of air concentration at high exposures, but not at the lower levels relevant to human exposures from Superfund site chemicals (32). The relationships that hold at these lower levels have yet to be elucidated. In addition, there is a great deal of variability in this coefficient among individuals, at least partly due to variations in blood chemistry (32). The sources of this variability need to be better characterized.

Physiological factors also affect absorption. For example, pulmonary absorption of some compounds is greatly increased by exercise (33). In addition, equilibrium between air and blood is often not achieved under these conditions. Another physiological factor, the percentage of body mass that is fat, has an influence; the higher the percent, the greater the absorption.

A two-compartment model can be used to describe pulmonary absorption. The parameters that need to be determined are inhaled concentration, rate of pulmonary ventilation, blood-to-air partition coefficient, venous blood concentration, and blood flow (31). Ventilation rate is an important factor that varies as a fractional power of body weight, so that children have a higher resting pulmonary ventilation rate per unit mass than adults. Ventilation rate also increases with exercise. Another factor of importance is the rate of metabolism of the absorbed compound. Thus, prediction of pulmonary uptake requires knowledge of a number of parameters and how these vary with factors such as exercise, body weight, and blood chemistry. In addition, the kinetics of the process need to be better characterized so that the applicability of equilibrium assumptions can be determined and deviations from equilibrium can be taken into account. More information will lead to decreased variability in assessing human exposure to airborne pollutants.

Personal Monitoring Data

Addressing the impact of human behavior on exposure to Superfund site chemicals requires much more information than is generally available. Issues involved in exposure assessment were addressed by P. Lioy (University of Medicine and Dentistry

of New Jersey). Occupational factors, recreational activities, in-home contaminants, and modes of transport are just some of the parameters that are needed to assess all possible pathways of exposure and thus evaluate the fraction that might be due to chemicals released from a Superfund site (34–37). To increase our knowledge of these parameters, more appropriate personal monitoring data and better biomarkers need to be developed. Once this is done, it may be possible to establish links between environmental parameters and these biological markers and thus use the latter more fully in exposure assessment.

A study that illustrates the type of information needed is one measuring exposure to volatile organics in bathing water (38). This research examined the relative importance of different routes of exposure to volatile organics in drinking water. The body burden of individuals taking a 10-min. shower with chloroform-spiked water was measured and found to be about 60% of the dose received from 2 L of drinking water containing chloroform at the same concentration. The amount absorbed was proportional to the time of exposure. Companion studies showed that about half of the dose was inhaled and half was absorbed through the skin.

Another study looked at the use of biomarkers to measure exposure to chromium from hazardous waste sites. Chromium VI is a very mobile substance and can seep through walls and floors of buildings, leading to significant levels in indoor air and on surfaces used by occupants. Thus, people living in the vicinity of chromium disposal sites can show complex patterns of exposure, including both indoor and outdoor components, that are difficult to quantitate from measured environmental levels. This study suggests that a better measure of chromium exposure may be gained by monitoring urine levels rather than determining ambient levels.

Another study focusing on human monitoring is an investigation of dietary exposure to PCBs from hazardous waste in Mohawk women and children in New York State. This was discussed by E. Fitzgerald (State University of New York at Albany). The study population is drawn from a community of about 10,000 Mohawk Indians who live at Akwesasne, a native American reservation along the St. Lawrence River. It is located near a General Motors Corporation waste disposal facility heavily contaminated with PCBs. In addition to environmental contamination from this operation, nearby Reynold's Metals and ALCOA facilities have also contributed to PCB releases into the environment.

As a result of these releases, PCBs have been found at significant levels in St. Lawrence River water and sediments. PCB contamination was also found in fish, turtles, and ducks, which are common food sources for the residents. To assess PCB exposure, lactating Mohawk women and their children are being studied. The research includes congener-specific analyses of breast milk, food sources, and environmental media. In addition, urinary levels of PCB congeners in both mothers and offspring will be measured and compared to breast milk levels. Interviews will be conducted in conjunction with these measures to estimate dietary, occupational, and residential exposure.

This study was begun in 1989 and has progressed through interviews of about 100 women and analysis of breast milk and maternal urine samples. Infant urine collection will begin soon. Mohawks who consume small quantities of contaminated

wildlife will be used as controls. The use of non-Indian populations as primary controls is being avoided to minimize the influence of genetic factors. Based on the size of the population and birth rate, it is expected that about 50 mother-child pairs will be enrolled in the study each year, through its conclusion in 1991. If exposure is documented, future studies focusing on infant growth and development will be conducted (39-43).

Wildlife as Monitors

In addition to direct studies of humans, investigations of wildlife can be performed and the results used as indicators of potential human exposure to chemicals present in Superfund sites (44). M. Hooper (Clemson University) discussed the use of wildlife as sentinels of exposure. To use such animals successfully, it is important to select species that are sensitive to the class of chemicals of concern, tolerant to experimental manipulation, exhibit wide geographical distribution, are present in statistically significant numbers, and have been well characterized biologically and ecologically.

Avian species fit many of these criteria and are commonly used as indicators. Examples include bobwhite quail, great blue herons, mallards, kestrels, hawks, starlings, and red-winged blackbirds. Those animals selected have been subjected to controlled short and long-term toxicity tests and exposure assessments to establish the relationship between exposure and dose. In addition to determining measures of acute toxicity, biochemical indicators have been assessed, as well as measures of teratology, growth rate, and fecundity. Dose is estimated in the field based on excreta, plasma, and tissue analyses; but these measures may be complicated by natural population variability and seasonal changes (45).

A specific example is the use of starlings to evaluate a terrestrial food web. Starlings are good sentinels because they nest in purposefully placed boxes, are tolerant to handling, are omnivorous, and are well characterized with respect to growth and biochemistry. Though the adults are not as sensitive as other species to many chemicals of concern, the nestlings have proven to be sensitive indicators of exposure and impact (46). Another example is the use of herons to evaluate bioavailability of contaminants because they eat frogs, fish, and small mammals. In this case, sampling of the chorio-allantoic membrane was used and was shown to predict within a factor of two the contaminant concentrations in whole eggs.

Although the utility of wildlife sentinels has been established, more research is needed to determine the appropriateness of specific animals, test methods, and end points. In addition, greater analytical sensitivity and improved biochemical assays will be needed for nondestructive estimation of dose in the field. Finally, more information is needed on the applicability of effects on one species to other species, including humans.

Case Studies

The preceding sections have focused on either environmental distribution and transformation or human behavior and monitoring. The strengths and limitations of models for movement within and among environmental compartments and within and among biota (including humans) were revealed in these sections.

However, a more complete understanding of the current status of exposure assessment can be gained from examining situations where a combination of environmental and biological models was employed. Following are three such situations.

Mercury in the Environment

T. Clarkson (University of Rochester) provided an assessment of human exposure to environmental mercury as an illustration of the importance of understanding environmental distribution and transformation in conjunction with human toxicokinetics [for detailed reviews, see Clarkson et al. (47) and WHO (48)]. There are two forms of mercury, inorganic and organic; and the organic form is generally of more toxicological concern because it is much more readily absorbed by the gastrointestinal tract. The organic form commonly occurs as the result of microbial transformation of inorganic mercury in aquatic systems, especially sediments. Thus, while the organic form is of ultimate significance, understanding the distribution and fate of its precursor, inorganic mercury, is critical in assessing potential exposure from hazardous waste sites.

Mercury metal readily enters the atmosphere, and is transported globally and converted to ionized forms before it is deposited. In addition, it is mobile in soil and so can move from waste sites to surface water. Once converted to organic forms, especially methylmercury, it can bioaccumulate in aquatic life and become biomagnified by over 1,000,000 times compared to water concentrations. Human intoxication through ingestion of methylmercury in fish has been documented around the globe with the most serious incident occurring in Minimata, Japan, resulting in many deaths.

The pharmacokinetics of methylmercury in humans is well understood; over 95% of the ingested dose is absorbed and elimination occurs with a half-life of about 50 days. The target site is the nervous system and symptoms include numbness, ataxia, and visual impairment. There is a latent period of 4-6 weeks, during which time irreversible damage can occur. Body burdens can be assessed with hair samples, and hair levels of 100 $\mu\text{g/g}$ seem to be the threshold for symptoms to appear in adults.

Fetal exposure to methylmercury can cause widespread central nervous system toxicity including effects on cell division and neuronal migration. The threshold in maternal hair for these effects is 10 $\mu\text{g/g}$ indicating that fetal populations are extremely sensitive to methylmercury effects and that toxicity can occur without concomitant effects in the mother. Human populations at most risk of mercury toxicity are those in which consumption of fish contaminated with methylmercury is heavy. Women of childbearing age are especially at risk due to the extreme sensitivity of the fetus.

Superfund Site Analysis: Air

The use of a particular modeling tool, air pathway analysis (APA), for assessing exposure and potential human health effects was described by D. Fingleton (Argonne National Laboratory) using a site near St. Louis, Missouri, as an example. APA is a systematic approach involving both modeling and monitoring to estimate exposures to individuals near waste sites (49). Air differs from other environmental media in that the migration times are much shorter, the impact area is larger, and mitigation of the potential consequences of a release into the atmosphere is much

more difficult. APA provides information that is useful for managing on-site worker health and safety as well as exposure to local populations. It also provides critical input for risk assessments and selection of remedial actions as well as development of emergency contingency plans.

To perform an APA, information is needed about sources (lagoons, leaking drums, etc.), fate and transport, and potentially affected populations. Monitoring can be used to establish air levels before and during remediation but such measures contain large uncertainties due to source variability and heterogeneity in concentrations of pollutants. Estimation of chemical transport is also difficult and calculated annual average estimates of ambient levels can vary over eight to nine orders of magnitude. This variability reflects uncertainties in the source terms, fate and transport models, and population distributions. Estimation of health effects on exposed populations is also uncertain (50,51). It is suggested that more realistic APAs can be achieved using a stochastic approach rather than a deterministic approach.

A site-specific application of APA was attempted at a Superfund site near St. Louis containing radioactive and chemical wastes (52,53). Successful application of APA is always difficult due to multiple sources, heterogeneous media, lack of site-specific meteorological information, poorly characterized contaminants, and on-site monitoring gaps. However, much more is known about this site than most others, so the uncertainty in the analysis was much smaller than usual.

However, as is commonly the case, behavioral information about local populations was limited. In the absence of such data, calculations were performed for two different exposure scenarios: passersby walking periodically at the fence line and trespassers. The results indicate that the cancer risk due to radiation was much greater than that due to chemicals for the passersby but not for the trespassers. For noncarcinogenic effects, exposure was much lower than the reference dose (RfD) for the passersby but comparable to the RfD for the trespassers. Although the absolute numbers calculated for this scenario contain some uncertainty, they represent a conservative estimate of the risk associated with the site, based on available data.

From this example, it is clear that more accurate and less uncertain exposure assessments depend on better and earlier input from the risk assessors. For example, the assessor can help define the purpose of the exposure assessment, e.g., short-term versus long-term effects. This type of input can contribute to better monitoring protocols and the collection of the most relevant data.

Superfund Site Analysis: Surface Water

Runoff or direct dumping of wastes into surface water is often a problem at Superfund sites. Models of fate and accumulation of chemicals in aquatic systems have been developed to describe such situations, and some have been tested directly at Superfund sites (54). One such study, performed at the Marathon Battery Site in Cold Spring, New York, was discussed by R. Thomann (Manhattan College). This facility is adjacent to a cove and the Hudson River estuary (55). The contaminant of concern is cadmium, and loading of this metal to surface water ranged from 6 to 18 metric tons annually over about a 20-year period ending in 1971.

Steady-state and time-variable models were developed to

describe movement between bodies of surface water (cove and estuary) and between sediments and surface water. The models incorporate advection, tidal dispersion, partitioning of cadmium to suspended and sediment solids, interstitial diffusion of cadmium, and net burial in the deep sediments. The approach follows that of Thomann and Di Toro (56). The models provide a method of determining contaminant concentrations and performing a mass balance. The time-variable model was applied to determine the past discharge history over a 25-year period as well as to predict future exposure levels. The ability of the model to extrapolate into the past was validated by using current data sources to model 1985 water column and sediment data for cadmium.

Using monitoring and modeling, it was possible to implicate consumption of blue crabs as the major source of human exposure to cadmium from this site. A simplified cadmium uptake model indicated that consumption of sediment cadmium and exposure to water column cadmium were important sources of cadmium in the crabs. Using the model, it was calculated that cadmium levels in crabs in the 1960s were such that consumption of reasonable amounts would have led to exceeding acceptable daily intake (ADI) levels by more than an order of magnitude. Current cadmium levels, although lower, would still lead to consumption of about twice the ADI. No epidemiological information is available to determine if adverse health effects resulted from the consumption of heavily contaminated crabs in the 1960s. Such information is critical for validating the use of site-specific models to estimate exposure levels in the past. The model is currently being further validated with sediment cadmium data up through 1989 to determine what sediment areas might be most effectively dredged to reduce current cadmium levels in the water column, sediment, and biota.

Summary

Assessing human exposure to chemicals from Superfund sites requires knowledge of basic physical, chemical, and biological processes occurring in the environment and specific information about the local environment and population in the vicinity of sites of interest. Although progress is being made in both areas, there is still a tremendous amount to be done. Participants at this meeting have identified several of the areas in need of greater understanding, and they are listed below.

- Movement of dissolved and volatile organics, especially NAPLs, in the subsurface environment. This includes study of the partitioning of compounds between NAPLs, air, water, and soil.
- Partitioning of volatilized chemicals between gaseous and aerosol components of the atmosphere. This includes understanding how these components influence both wet and dry deposition.
- Long-term movement from sediments into biota and how these affect chronic toxicity to sediment biota.
- Broad validation of PBPK models describing partitioning of compounds from sediment and water into fish.
- Reactions of chemicals sorbed to atmospheric particles. This includes application of laboratory models to real and varied atmospheric conditions.
- Interactions between biotic and abiotic transformations in soil and sediment.

- Applicability of physiological pharmacokinetic models developed in laboratory studies of experimental animals and clinical investigations of humans to environmental chemicals, concentrations, and routes of exposure in humans.
- Use of human and wildlife behavioral and biomonitoring information to estimate exposure. This includes better understanding of human variability and the applicability of information gathered from particular wildlife species.

To successfully address these gaps in our knowledge, much more analytical data must be collected. These data are needed to better define the heterogeneity found in environmental situations, to provide the raw material necessary for the development of models, and to provide the data critical to testing and refining these models. In addition, more and better data need to be collected at Superfund sites to reduce uncertainties when applying the models that have been developed.

Appendix

Assessment of Human Exposures to Chemicals from Superfund Sites: Program

Session I

Chairperson: William Thilly, Massachusetts Institute of Technology
Multimedia transport

Yoram Cohen, University of California at Los Angeles

Physiologic pharmacokinetic modeling of pollutant chemicals

Robert Dedrick, National Institutes of Health

Exposure evaluation: a multidimensional approach

Paul Liroy, University of Medicine and Dentistry of New Jersey

Session II

Chairperson: Max Costa, New York University

Biomarkers of exposure. Meeting Report

Martyn Smith, University of California at Berkeley

Modeling fate and accumulation of chemicals in aquatic systems

Robert Thomann, Manhattan College

Predicting bioavailability and toxicity of metals in sediments

Dominic Di Toro, Manhattan College

Uptake of pollutants in fish

Russell Erickson, Environmental Protection Agency

Dietary exposure to PCBs from hazardous waste in Mohawk women and children in New York State

Edward Fitzgerald, State University of New York at Albany

Human exposure to mercury

Thomas Clarkson, University of Rochester

Retention of contaminants in soil

Thomas Voice, Michigan State University

Session III

Chairperson: Roy Albert, University of Cincinnati

Degradation of soil contaminants

Timothy Vogel, University of Michigan

Biodegradation of pollutants. Meeting Report

Steven Aust, Utah State University

Wildlife as sentinels of exposure

Michael Hooper, Clemson University

Atmospheric fate and transport

Haromi Niki, York University

Air pathways analysis of health risks for a Superfund site

Donald Fingleton, Argonne National Laboratory

Factors controlling pulmonary absorption of chemicals

Michael Morgan, University of Washington

Information gaps in exposure analysis

Lawrence J. Fischer, Michigan State University

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