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State of Research: Environmental Pathways and Food Chain Transfer

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Data on the chemistry of biologically active components of petroleum, synthetic fuel oils, certain metal elements and pesticides provide valuable generic information needed for predicting the long-term fate of buried waste constituents and their likelihood of entering food chains. Components of such complex mixtures partition between solid and solution phases, influencing their mobility, volatility and susceptibility to microbial transformation. Estimating health hazards from indirect exposures to organic chemicals involves an ecosystem's approach to understanding the unique behavior of complex mixtures. Metabolism by microbial organisms fundamentally alters these complex mixtures as they move through food chains. Pathway modeling of organic chemicals must consider the nature and magnitude of food chain transfers to predict biological risk where metabolites may become more toxic than the parent compound. To obtain predictions, major areas are identified where data acquisition is essential to extend our radiological modeling experience to the field of organic chemical contamination.

Introduction

The indirect hazards of energy-related products to humans may result from long-term fate and distribution of many organic compounds in terrestrial and aquatic ecosystems. Contamination reaching people through environmental pathways involves many different organisms (food chains) and chemical processes of a peculiarly environmental kind that need to be better understood. The chemical processes include metabolism in lower life forms, abiotic (geochemical) transformations, and microbial conversions any of which may produce more or less hazardous chemical forms. This review considers the state of the research on metabolism, how it relates to food chain and environmental transfer processes, and the modeling of pathways involved in these processes.

Polycyclic organic matter (POM) is a type of energy residual from pyrolytic, combustion and other fossil fueled energy sources, which has been found widely distributed in water, soil, air, plant tissues, and animal tissues (*1*). As discussed herein, recent research on complex mixtures containing POM indicate that their POM content, *per se,* may not be harmful to humans. Rather, specific polycyclic hydrocarbon compounds contained in the POM mixtures may accumulate with differing degrees of toxicity depending on the situation.

Petroleum, waste oils, shale oil and coal liquids show substantial differences in regard to the potential carcinogenic activity of POMs contained therein, as judged by mutagenicity (*2,3*). Also, levels at which mutagenic activity can be demonstrated are well below chronic or acute toxicity levels (*4*). In considering indirect hazards to humans, data on mutagenic activity are probably more important than data on acute toxicity. In considering ecological effects, or the well-being of ecosystems, chronic toxicity data cannot be ignored (*5*), but that is not within the scope of this review.

It is not possible to study all of the organic

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products generated by various fossil energy technologies, and for this reason representative compounds of chemicals need to be selected for in-depth studies. These organic products constitute complex mixtures with unique properties that present problems in measuring and predicting biological effects. The greatest challenge of the next decade will be to establish the principal features of the interactions of the chemically complex mixtures with organisms and their abiotic environment. To reach a general understanding on these interaction features will probably be as complex an undertaking as was our approach to radiation biology 80 years ago.

Our radiological experience is valuable in trying to inter-relate a complicated pattern of different chemicals, different organisms, and different environmental processes. The sheer complexity makes a formidable task of studying and trying to describe the result of indirect human exposure to chemicals through environmental pathways that include food chains. Fifteen years of research on polycyclic aromatic hydrocarbons (PAH) have provided a strong head start in understanding biological uptake and retention of at least one class of organic compounds (*4*). Intensive work undertaken over the past five years on coal-liquids has provided some further insights particularly on the characteristics and environmental behavior of other complex mixtures (*5,6*). However, overall, there are many gaps to be filled in the data base before models can confidently predict food chain hazards from the more toxic complex mixtures like coal liquids (*7*).

This review is not intended to be comprehensive in any one area. Rather, it is written to illustrate key points relative to food chain transfers and waste residuals; and, to spotlight major areas of uncertainty requiring research. A variety of polycyclic hydrocarbon compounds will be discussed. (Fig. 1).

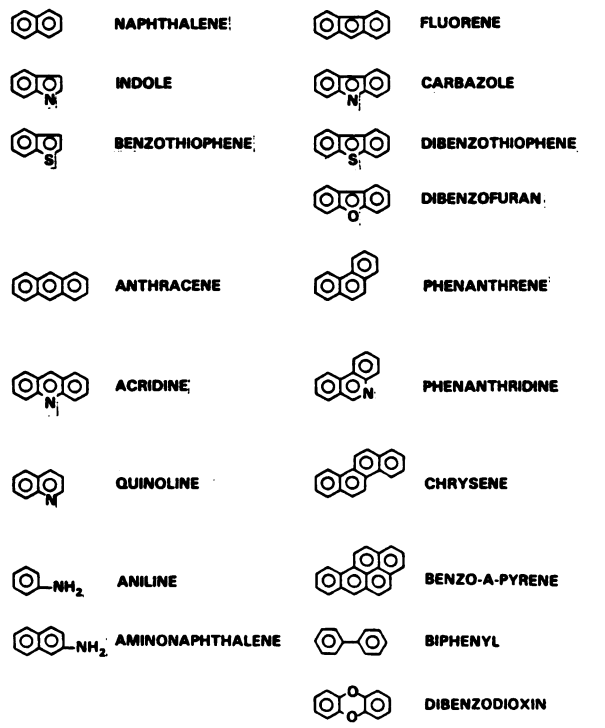

FIGURE 1. Schematic drawing of compounds discussed in text. Derivative compounds may be found in Weast and Astle (*8*).

Modeling Perspective

The experience with radiological models used in estimating indirect exposures has been developed over a long period of time, and it has led to a well-defined conceptual approach needed to deal with the various environmental processes and pathways (*9*). Problems in applying such models to a chemical contamination situation have been reviewed elsewhere (*10,11*), and recent attempts have been made to apply radiological modeling to organic chemical contaminants (*12*). At the present time, such an approach is only limitedly useful. But it can be used to determine whether adequate data bases exist, in situations where the number of pathways and "sinks," i.e., where a compound can be accumulated, are too numerous to track (Fig. 2). The model helps us answer such questions as: Do we have at hand any source (emission) data? Is the source a particulate or gas or both? Do we have hydrologic and atmospheric dispersal data? Do we have sorption or microbial conversion data for soil or sediment? We must look at the entire ecosystem in considering exposure situations involving food chains, and a significant aspect in developing the data bases is acquiring information on key environmental processes affecting entry or subsequent transformation of the chemicals in question.

The radiological modeling approach taught us to get systematic physical and chemical data on source "terms" and critical pathways. Source terms include particle size (airborne), physical state, chemical form, simultaneous presence of complexing agents or solubilizers, and release rates. A field contamination situation involves the transfer of pollutant from soil to plant to herbivore, which makes it a critical pathway to people. The source terms fundamentally control biological availability at the plant leaf, root or

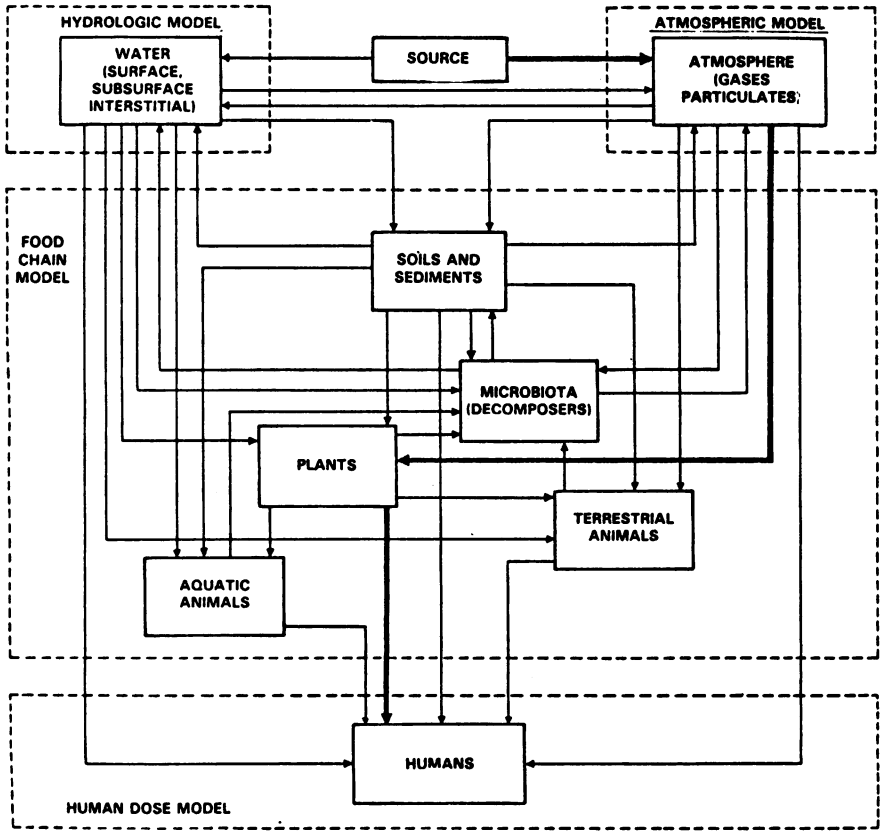

FIGURE 2. Modeling contaminant transfers from the environment to man—a conceptual framework based on radiological experience. The model shows major compartments, various biotic and abiotic processes of transfer between compartments, and four major submodels on which the computer codes are based. Illustrated is a critical pathway for transfer from air to plant to man. In the pathway shown, e.g., by boldface arrows, substantial problems arise in application to organic compounds (see text). Based on Vaughan et al. (*9*).

soil interface; failure to take them into account can change uptake determinations by several orders of magnitude (*9*). In the chain of a critical pathway, metabolism by a food chain element can significantly alter contaminant uptake at subsequent steps. Only a few data are available on the chemical form taken up and transferred between trophic levels or between soil and plant. Such data are scarce even for those elements where chemical change is known to be important (*13,14*). PAH compounds have been better studied but data on metabolites besides those discussed in this report are not plentiful. The greater concern over metabolites is now clearly recognized, however (*1,15*). Microbial metabolism of PAH in petroleum fractions has been rather well reviewed (*16–18*), but the extent to which soil or sediment metabolites would impact food chains is not clear.

Developing Chemical Exposure Models

The best known and most widely utilized exposure model derives from radiological experience. Current topics of concern for the radiological model are shown in Table 1 and have been treated fully elsewhere (*9*). Other features need to be considered in developing chemical exposure data bases that might ultimately be useful for modeling purposes. Source factors and passive transport processes remain equally important for radiological and chemical models. However, mobilization and uptake, and biological availability, are in some cases complicated by metabolism and the unique properties of complex mixtures.

Uptake and possibly metabolism can be markedly altered by processes that occur upon

Table 1. Comparing information data bases.

Radiological type models	Organic chemical type models
Source factors	Source factors
Dispersal mechanisms (hydrological, atmospheric)	Dispersal mechanisms
—	Complex mixture behavior
Mobilization and uptake	Mobilization and uptake
—	Metabolism
Critical (environmental) pathways	Critical (environmental) pathways
Estimation error	Estimation error

deposition. Atmospheric particulate sorption may be the most significant process for hydrocarbon contamination from airborne sources (*19*) as it is for radioelement contamination (*20*). Complex mixtures at the source often introduce further complications that are not recognized in radiological modeling.

Mobilization and uptake depend on transport mechanisms at the cellular level, and these mechanisms have not been well studied for complex ring-type compounds. Specific compounds or their metabolites generally require an enzyme-mediated step for uptake. Subsequently, they may be accumulated by the organism inadvertently, either blocking normal metabolic activity, as happens with certain pesticides (*21*), or enhancing biochemical activity that has high likelihood of causing injury, as discussed below. Mobilization and uptake mechanisms are not well characterized as to the specific chemical forms they accept. Also, the organic chemical forms that would modify metabolism or accumulation have not been systematically studied.

Radioelement exposure models generally assume that an element moves through environmental pathways largely unaffected by metabolism. This has been always a questionable assumption, with the consequence that in soil systems involving green plant, gross error ranges from 1 to 10^{-3} for soil to plant transfer ratios can be found. The single most important model segment affecting these large errors has been the soil submodel, for both radioelements (*9*) and metals (*22*). The focus on delineating soil processes that control biological availability of nonnutrient compounds like certain radioelements is relatively recent (*23–25*). Data on microbial degradation of organic compounds are plentiful but have not yet been brought to a sharp focus as regards controlling variables (*16,18,26,27*).

Persistence holds the key to appropriate exposure models for chemical contaminants. A persistent compound is likely to move through several links of a food chain system and possibly accumulate at the topmost levels (biomagnify). In animals, this usually happens when the enzyme systems are lacking to efficiently metabolize a particular organic compound, such that it accumulates in fatty tissue, like hexachlorobenzene (*28*). In these instances, solubilization will determine uptake, but as discussed below, solubilization in complex mixtures usually depends on several processes, e.g., partitioning, hydrotropy, emulsification, in addition to chemical solubility. Alternatively, in plants—and less typically in animals—accumulation in a particular organ may depend on ligand formation that is in turn dependent on the bonding characteristics of a particular element, like cadmium (*29*). Activity in other compounds, like PAH, will be related to overall molecular configuration rather than to the bonding characteristics of a particular element within the molecule and the activity is likely to be significantly altered by the fate of metabolic conversion products. Between these extremes, there lies a distribution of chemical compound behaviors.

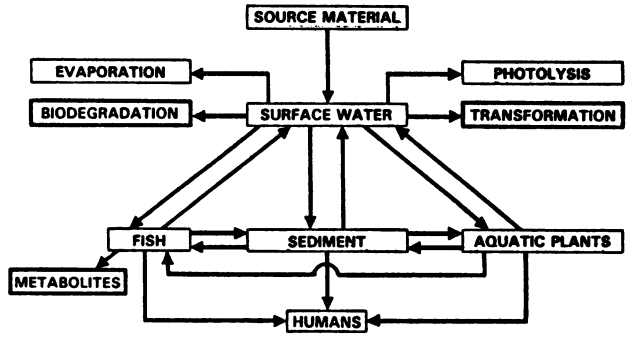

FIGURE 3. Typical aquatic pathways exposure model. The model assumes steady state dynamic equilibrium with small losses from the system and makeup from the source. For many organic compounds, metabolites may not leave the system and (microbial) degradation is likely to be partial leading to accumulations of other metabolites. Hazard evaluation must incorporate a consideration of metabolites potentially more toxic than the parent compound. From Aaberg et al. (*31*).

Assumptions of a modified steady-state dynamic system are implicit in current chemical modeling attempts (*30,31*) as shown in Figure 3.

Associated with such models are efforts to systematize bioconcentration factors (BCFs) for a wide variety of compounds based on octanol–water partitioning coefficients (*32,33*). For chemicals that are being transported and redistributed between solid, liquid and gas phases, including biota, the steady-state approach may be of interest; however, it does ignore metabolism, and present model development is heavily weighted toward chlorinated and other hydrocarbon classes of low metabolizability that persist in fatty tissue (*34,35*). With the present unsettled state of the art, it would seem unwise to force most data acquisition and its parameterization according to the implicit assumptions of steady-state kinetic models. This is particularly true because metabolism and transformation reactions are not well understood and cannot be explicitly factored into such models.

Abiotic Transformation Reactions

Atmospheric and hydrological dispersal sometimes are looked upon as passive physical processes; however, such processes are subject to chemical modification through transformation reactions. These reactions have not received the attention they need for chemical modelling purposes, and they may also affect metabolism. Effluent materials frequently are altered chemically through abiotic reactions, which can be caused by elevated temperature and/or free radicals present in atmospheric plumes, by salinity or other changes in hydrologic plumes, or by geochemistry in soil systems. Other factors that may alter chemicals include photooxidation, volatilization and various particle physicochemical or geochemical transformations (*17,36*). In actual soil systems, geochemical processes are often inextricably bound up with microbial metabolism (*37*). It may be important to delineate the truly geochemical processes, but this has not yet been fully accomplished in soil systems.

Most atmospheric PAH compounds are associated with particulate surfaces (*1*), but the reactions of surface-associated molecules are only beginning to be investigated (Fig. 4). For example, certain PAH sorbed to fly ash particles were found to be stabilized against photochemically induced decomposition; the compounds included benzo(a)pyrene, methylbenzanthracene, acridine, fluoranthene and phenanthrene (*40*). Others were found, in the same study, spontaneously to decompose when sorbed to the fly ash; these compounds generally contained a benzylic nucleus, like fluorene, benzofluorene, or azafluorene. Quinones and ketones resulted from decomposition.

Evidently, particle sorption of PAH takes place after fly ash escapes the stack, such that air concentrations of particulate PAH will be grossly underestimated by stack sampling (*19*). Atmospheric gases like NO_2 can produce compounds with increased or perhaps previously unobserved mutagenic activity. For example, NO_2 at from 1 to

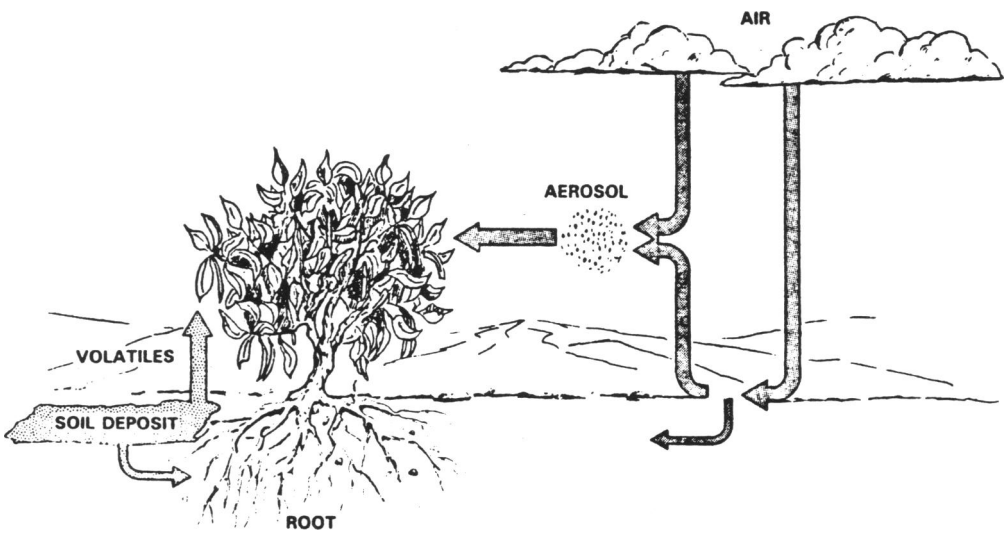

FIGURE 4. Plant uptake of polyaromatic hydrocarbons (PAH). Most atmospheric PAH are sorbed to particles less than 2 μm in size, where they may be stabilized and/or nitrosated. Foliar uptake often greatly exceeds root uptake from soil. PCB sorbs to leaves by volatilization from soil with little or no root absorption. Based on Fries and Marrow (*38*), Lewtas et al. (*39*) and Cataldo and Vaughan (*20*).

100 ppm caused formation of the more mutagenic nitro derivatives of benzo(a)pyrene and perylene sorbed to fly ash (*41,42*). SO_2 tests were ambiguous.

The importance of these observations for people exposed to pathways involving green plants cannot be overlooked. There seem to be no comparable data on organic compounds other than the PAH.

Somewhat analogous events are known to occur in water columns, particularly affecting materials sorbed to suspended particulates, and they have been reviewed elsewhere (*17*); examples cited include photooxidation, sorption on mineral colloids and oil droplets, and other surface-active factors. The likelihood of these events points to a serious obstacle in extrapolating from a laboratory observation to a field prediction.

Volatilization

Volatilization is a complicating factor, from the standpont of environmental pathway modeling, which may bias estimations of the transfer of pollutant from sediment to detrital organism, or from soil to plant root (see Fig. 4). Lower molecular weight PAH and some chlorinated hydrocarbons have an appreciable vapor pressure (*4,43*), which suggests that their volatility in solution may be as significant as their volatility in air. How important this factor may be in biasing the entry of such compounds into the pathways shown in Figure 2 is not easy to ascertain. It appears to be an important factor in terrestrial plant uptake from soil surfaces (*44*), but it is probably of minor importance in detrital organism uptake from benthic sediments contaminated by accidental spills. However, volatility may greatly alter the distribution of compounds that organisms may be exposed to in surface spills of petroleum (*17*) or other products.

Features of Complex Mixtures

Petroleum typifies the measurement problem in dealing with mixtures of many hundred thousands of organic compounds, each of which shows different solubility, oil-water partitioning, particle-water partitioning, emulsifying, toxicological and other properties (*4*). Coal synthetic fuels, shale oil, and the organic fraction of industrial wastewaters or raw river water present similar problems (*45–49*). Practicable approaches to measurement focus on solubility, octanol–water partitioning, and, particularly, on determining exposure spectrum, based on the measurement of reference compounds.

During an interval of exposure to a complex mixture, particularly in an environmental situation, an organism experiences a continually shifting spectrum of individual compounds (Fig. 5). This has been demonstrated with petroleum and coal synthetic oils (*5,50–52*), where fairly elaborate procedures have had to be developed to assure consistently reproducible responses; the procedures required periodic determinations of the concentrations of the desired reference compounds. In determinations of how an organism handles a particular compound metabolically, in the presence of a complex mixture, the experiment can be easily confounded if opportunities exist either for partitioning or for blocking. Hence, for either predictive purposes or for control of experimental procedures, it is essential to chemically validate the exposure conditions. Cur-

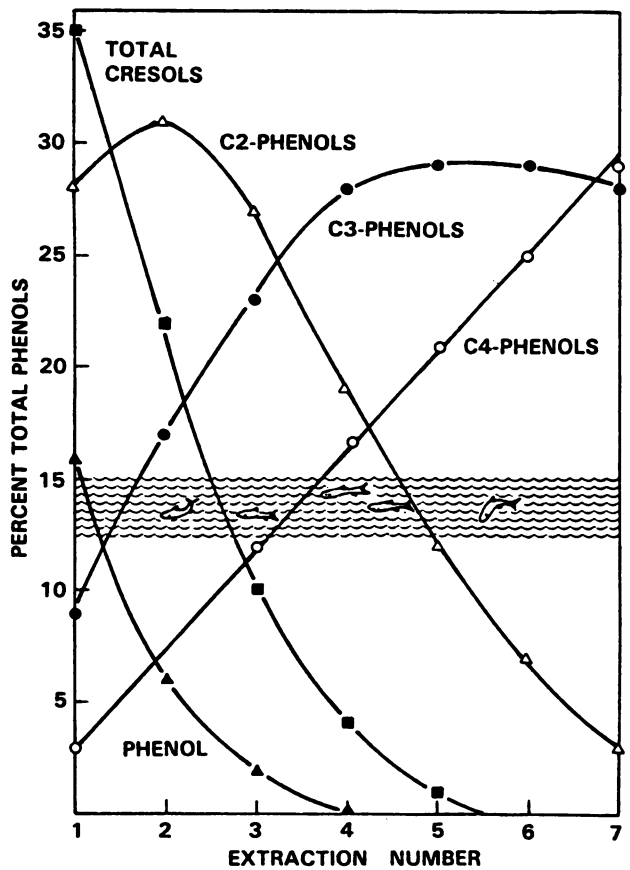

FIGURE 5. Successive extractions of a water soluble fraction of coal liquid represent the time course of partitioning of a mixture of individual phenols and other compounds. In nature, fish swimming through a contaminated zone will be exposed to a continually shifting spectrum of compounds represented by these phenolic compounds of varying molecular weight. Based on Strand and Vaughan (*5*).

rent development in chemical fractionation and GC-MS methods make validation practicable.

Solubilization

Solubilization for chemical modeling of food chains refers to more than chemically defined solubility. In complex mixtures, other processes that need to be considered are emulsification, hydrotropy and partitioning on suspended solids. Any of these processes may effectively bring materials into contact with the uptake surfaces of biota. Natural and waste waters often contain significant amounts of other dissolved and colloidal organic materials capable of incorporating oily compounds into the micelles of surface active molecular aggregates (*4*); humic acids, fulvic acids and other common plant biodegradation products provide good examples of such solubilizers (*17*). Hydrotropy is the enhanced solubility of organic compounds in the presence of other organic compounds that are not themselves colloidal (*4*).

Partitioning

Fairly extensive literature exists on aqueous solubility and octanol-water partitioning of numerous organic compounds (*4,17,33,53,54*). Octanol partitioning gives an estimate of the potential for accumulation in lipids, and it generally correlates well with the relatively simple bioconcentration test using daphnids (tissue concentration/water concentration). However, as compared to the daphnid test, the same approach in fish has been found greatly to over estimate bioconcentration of azaarenes (*55*), because fish rapidly metabolize this particular organic compound class. Thus, only experimental measurement can establish the actual degree of bioconcentration (or biomagnification through higher trophic levels.)

If metabolism of one compound in an organism is known, partitioning data may be useful for comparative estimation of fat accumulation among several similar compounds. For example, the progressive addition of from one to three methyl groups to naphthalene decreased its solubility in water by an order of magnitude (*56*), and an increase in molecular weight (MW) among similar azaarenes (benzacridine, quinoline and isoquinoline) caused a proportional decrease in solubility and increase in fat accumulation (*55*). Sulfur or nitrogen heterocycle analogs of PAH such as anthracene or benzanthracene show increased polarity and, as a consequence, possibly greater water solubility (*57,58*). However, it is not clear that these heterocycles follow similar metabolic pathways, so they should not be compared to PAH based on partitioning data alone.

In soil systems, as compared to aquatic systems, much less information is available. Partitioning between the soil water and the solid fraction is known (*59*), and it is highly correlated with the soil organic fraction in both soils and sediments (*4,60*). An active microfloral metabolism is characteristic of most soils and sediments (*17,27*). For similar reasons the utility of partitioning data may be no more or less in soil systems than it seems to be for aquatic systems.

Blocking and Synergism

Probably with most complex mixtures, as with coal liquids, toxic activities (including genotoxic activity) of constituent compounds are not simply additive. From a modeling point of view, this feature is quite unlike radiation or heavy metal accumulation. In complex mixtures from coal liquids, both significant blocking and synergistic activities have been found recently (*61,62*). Also, the mutagenicity of pure benzo(a)pyrene (BAP) can be progressively inhibited by adding back incremental portions of crude fraction of coal liquid, based on the Ames assay (Fig. 6). When the data are corrected for fractional weight in the crude mixture, comparable results can be demonstrated on distillate cuts of coal liquids in the 700–800°F boiling point range. An entirely differ-

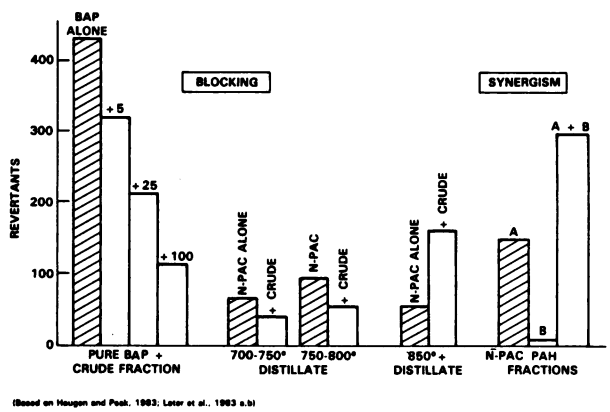

FIGURE 6. Blocking and synergism. When crude fractions are added back to benzo(a)pyrene (BAP), its mutagenic action is progressively suppressed. Distillate fractions below 850° F showed similar findings. The distillate fractions are enriched in nitrogen containing polyaromatic compounds (N-PAC). Mutagenic activity of a highly purified N-PAC fraction was doubled when a nonmutagenic PAH fraction was added back (data are corrected for weight fraction and portrayed at 1/10 of the ordinate). Synergism was seen similarly for distillate cuts above 850°F. Data taken from Later et al. (*63,64*).

ent result occurs above 850°F; the distillate cuts contain high proportions of nitrogen containing polyaromatic compounds (N-PAC) that are more highly mutagenic than BAP. When crudes are added back to these particular fractions, their mutagenic activity is nearly tripled, rather than blocked. This striking synergism also has been confirmed with highly purified N-PAC fractions activated by otherwise inactive PAH fractions. In determining toxic response of organisms, it therefore becomes essential to couple specific compound testing with chemical fractionation and testing of the mixture, not only for determining mutagenic activity but also for determining other types of biological activity.

Significant blocking and synergistic activities have also been detected in complex mixtures from shale oil (*65, 66*). Raw shale oil, like coal liquids, differs from most crude petroleums in having comparatively high concentrations of basic (nitrogen-containing) and phenolic compounds that are not present in petroleum oils.

Table 2. Risk assessment units proposed for complex mixtures.[a]

Number	Category	Description
1	Carbon monoxide	CO
2	Sulfur oxides	SO_x
3	Nitrogen oxides	NO_x
4	Acid gases	H_2O, HCN
5	Alkaline gases	NH_3
6	Hydrocarbon gases	Methane through butanes, acetylene, ethene through butenes; C_1-C_4 alkanes, alkenes, alkynes and cyclo compounds; bp <~20°C
7	Formaldehyde	CHO
8	Volatile organochlorines	To bp ~120°C; CH_2Cl_2, $CHCl_3$, CCl_4
9	Volatile carboxylic acids	To bp ~120°C; formic and acetic acids only
10	Volatile O&S heterocyclics	To bp ~120°C; furan, THF, thiophene
11	Volatile N heterocyclics	To bp ~150°C; pyridine, piperidine, pyrrolidine, alkyl pyridines
12	Benzene	Benzene
13	Aliphatic/alcyclic	C_5 (bp ~40°C) and greater; paraffins, olefins, cyclocompounds, terpenoids, waxes, hydroaromatics
14	Mono/diaromatic hydrocarbons (excluding benzene)	Toluene, xylenes, naphthalenes, biphenyls, alkyl derivatives
15	Polycyclic aromatic hydrocarbons	Three rings and greater; anthracene, BaA, BaP, alkyl derivatives
16	Aliphatic amines (excluding *N*-heterocyclics)	Primary, secondary and tertiary nonheterocyclic nitrogen, $MeNH_2$, DiMeNH, TriMeN
17	Aromatic amines (excluding *N*-heterocyclics)	Anilines, naphthylamines, amino pyrenes; nonheterocyclic nitrogen
18	Alkaline nitrogen heterocyclics ["azaarenes"] (excluding "volatiles")	Quinolines, acridines, benzacridines; excluding pyridines
19	Neutral N, O, S heterocyclics (excluding "volatiles")	Indoles, carbazoles, benzofurans, dibenzothiophenes
20	Carboxylic acids (excluding "volatiles")	Butyric, benzoic, phthalic, stearic
21	Phenols	Phenol, cresols, catechol, resorcinol
22	Aldehydes and ketones ["carbonyls"] (excluding formaldehyde)	Acetaldehyde, acrolein, acetone, benzaldehyde
23	Nonheterocyclic organo sulfur	Mercaptans, sulfides, disulfides, thiophenols, CS_2
24	Alcohols	Methanol, ethanol
25	Nitroaromatics	Nitrobenzenes, nitropyrenes
26	Esters	Acetates, phthalates, formates
27	Amides	Acetamide, formamide, benzamides
28	Nitriles	Acrylonitrile, acetonitrile
29	Tars	
30	Respirable particles	
31	Arsenic	As, all forms
32	Mercury	Hg, all forms
33	Nickel	Ni, all forms
34	Cadmium	Cd, all forms
35	Lead	Pb, all forms
36	Other trace elements	
37	Radioactive materials	Ra-226
38	Other remaining materials	

[a]Based on Moghissi and Foley (*45*); Moghissi, personal communication.

Criteria for Reference Compound Selection

Given the extraordinary number of individual compounds in complex mixtures here being discussed, their widely differing biological activities, and rather limited data, a selection of reference compounds for intensive study affords the only practicable way to commence developing a useful data base. EPA is developing a list of risk assessment units (Table 2) which attempts to classify compounds for this purpose (*45*).

This concept is likely to be refined by EPA because recent research findings reveal additional problems (A. Moghissi, personal communication). For example, benzo(a)pyrene, in group 15 (Table 2) was selected originally because of its potential carcinogenicity when administered as a single compound. However, as discussed earlier, its carcinogenic activity can be either blocked or potentiated in the presence of other constituents. Such findings illustrate a major problem in applying conventional toxicological criteria to a complex mixture. Other problems concern the biological availability and metabolic significance of particular compounds in a risk assessment unit.

In selecting reference compounds, different criteria sometimes may be desirable; high octanol partitioning may be considered where interest focuses on fat accumulation (*32,53*). It is not clear that fat accumulators are necessarily more carcinogenic than compounds showing lower octanol water partitioning coefficients, but fat accumulation may be very significant for food chain evaluation. A reference compound may also be selected because its solubility characteristics conservatively represent a wide range of other compounds. Dimethylnaphthalenes have been used, for example, to represent the water-soluble fraction of petroleum that contains toxic, volatile compounds that are otherwise difficult to measure (*50,51,67*). A reference compound might also be selected because its molecular configuration uniquely biases metabolism towards a metabolic pathway of interest, as do those strongly carcinogenic compounds that form bay region diol epoxides (*68*).

Chemistry research continues to emphasize compound classes based on fractionation techniques. Earlier work tended toward biologically arbitrary classification (*69*), but recent studies in which fractionation classes are guided by mutagenesis testing appear to be a promising development (*64,70,71*). *N*-Containing basic fractions seem to be particularly important in regard to mutagenesis for products from the new synfuels technologies.

There are other noxious properties to be considered besides the carcinogenic potential; e.g., chemical avoidance (*72*), acute or chronic toxicity of compounds that may effect food base organisms (*5,73*) and tainting of foodstuffs by compounds causing off-flavors (*74*). Clearly, the criteria to be considered in selecting reference compounds are multiple. No current consensus exists on how best to go about making that selection.

Metabolism

Of the POM taken up by most higher organisms, one–half or more partitions into their lipid constituents, and the smaller fraction enters their metabolic pool (Fig. 7). Long–term material may be recalled from lipid storage and metabolized. The PAH compounds, the biphenyls, and the heterocyclic analogs of PAH are to some extent metabolized by all forms of life, since the basic enzyme systems necessary to metabolize these compounds have been found in microbial populations, plants, insects, fish and mammals (*76,77*). The MFO system, on which the first phase of metabolism depends, is also inducible in mammalian (*16*), microbial (*17*) and fish species (*78,79*). Inducibility enables some aquatic ecosystems to adapt to chronic exposure to petroleum, and to adapt without selective loss of organisms from the

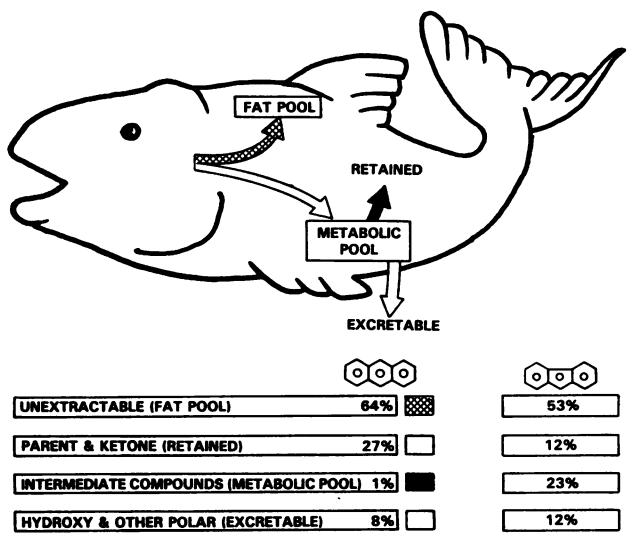

FIGURE 7. Relative distribution of metabolites for two PAH compounds. Fluorene is more readily metabolized than anthracene, with a comparatively larger proportion of excretable, polar metabolites and a lower proportion of unextractable compound (presumably parent compound sorbed into fat). Metabolites are listed in order of increasing polarity on thin layer chromatographs. Based on Lu et al. (*75*).

system (*49,80*). There is also some degree of substrate specificity as to the MFO induced, judging from petroleum experiments with fish (*81*); however, the range of specificities does not seem to have been delineated for any MFO system studied to date. Conjugation reactions, in the second phase of metabolism, are also common to most life forms. Reflecting their evolutionary linkage, higher plants, animals and microorganisms generally metabolize PAH by similar pathways, and interphyletic differences are thought to be more closely related to differences in organ circulation, metabolic efficiency, excretory efficiency, or predominant sites of metabolism (*77*).

Unlike radiological dose, degree of mutagenicity or carcinogenicity may not linearly correlate with the extent of accumulation of an organic hydrocarbon. Some compounds may accumulate in tissues passively with little or no injury potential. For a hydrocarbon to be mutagenic, it must be partially metabolizable, or at least be able to enter transport pathways and block normal enzymatic processes. Particularly in the case of ring–type structures, the possibility exists that conversion products of metabolism may be more hazardous than the parent compound. This has been shown, e.g., for genotoxicity in fish, although the compounds in waste oil were not identified (*2*). If toxic metabolites are produced, they warrant systematic evaluation as to their fate in the organism and likelihood of passage up a food chain.

Metabolic pathways include oxidation, reduction, hydrolysis, conjugation, acylation, alkylation, and ring cleavage. Unfortunately, however, the range of reactions and compounds of interest is so diverse that in many instances little information is available on the relative importance of particular pathways for a given compound and organism. Thus it is difficult to predict the predominant metabolites and their toxicities in relation to the parent compound. Most of our knowledge about the metabolism of the more complex organic compounds comes from laboratory studies on microorganisms (*16,18*). Very little has been done with controlled ecosystem enclosures and higher life forms. Efficiency of metabolism of branched versus straight–chain alkanes and of substituted versus simpler aromatics probably explains the "biological fractionation" of oils sometimes reported (*52*).

For illustrative purposes it is useful to compare compounds at two extremes of the spectrum: (1) PAH of low MW, like naphthalene, which are metabolized significantly and not appreciably bioconcentrated, and (2) compounds like the chlorinated biphenyls, which are only slightly metabolized, bioconcentrated, and biomagnified in food chains. In general, chlorination severely reduces the metabolizability of most organic compounds, including simple rings, heterocycles and PAH, as compared to the nonchlorinated analogs (*28*). The increased fat accumulation more or less correlates to degree of chlorination (*33*). This approach to the question of metabolizability may have predictive value, but it cannot be considered without close evaluation of the environmental behavior of metabolic products.

Polychlorinated Biphenyls (PCBs)

PCBs accumulate in fatty tissue of aquatic animals directly from water (*82*) and in the fatty tissue of other animals by ingestion of contaminated foodstuffs (*83*). While accumulation occurs, these compounds are not appreciably metabolized. Mono-, di- and trichlorobiphenyls are metabolized to phenolic compounds, as are tetrachloro derivatives, but more than four chlorinated positions on the molecule blocks metabolism in most microbes and all higher life forms (*28*). In mammals, a phenolic metabolite four or five times more toxic than the parent tetrachlorobiphenyl has been reported (*84*). In a natural assemblage of microbes, the selective biodegradation of monochloro compounds proceeded, while dichloro compounds also present in the commercial mixture were relatively enriched (*85*). Thus, congeners of higher chlorination degree will accumulate in sediments with possibly an enhanced likelihood of adverse metabolites being formed (if they are at all metabolized).

As regards the metabolically active portion, fish, birds and mammals, in that order, are increasingly able to convert biphenyls via the mixed function oxidase (MFO) enzyme system in liver (*28*). Aerobic bacteria are also capable of metabolizing PCB, but they do not do so appreciably in presence of other carbon sources; and, anaerobic bacteria do not degrade PCBs (*86*). Among vegetable crops, metabolism is minimal. In carrots about 97% of the PCB is on the peel and evidently not metabolized, except for a trivial fraction (*44*); this seems to be true of other leafy crops (*87*).

Polychlorinated Dibenzofurans (PCDF)

These compounds occur as impurities in commercial PCBs. They are structurally similar to the dioxins and were detected in the Pacific

Northwest's Puget Sound in a special study by NOAA (*28*). Practically no metabolic data are available for PCDF, but they are considered not to be metabolized to any appreciable extent (*28*). Potentially, they should biomagnify through food chains, by solubilization in fat deposits like the PCBs. Little is known about the toxicity of these compounds, and work should be undertaken to delineate their toxicities in relation to other known carcinogens. The structurally similar chlorinated dioxins, for example, are among the most toxic substances known, are fat accumulators and are not generally degradable microbially. Nonchlorinated compounds of this type which occur as by-products of coal conversion are oxygen heterocycles structurally similar to dibenzothiophene or carbazole (*75*). Whether they are more or less toxic than PCDF also is not known.

Polycyclic Aromatic Hydrocarbons (PAH)

These fairly intensively studied compounds constitute a very diverse class (*1*). Intermediary products of their metabolism may be more toxic than the parent compound. This happens because the hydroxylated products are more water soluble (*15*) and, for some of the PAH compounds, the diol epoxides formed in the normal course of metabolism will be uniquely carcinogenic because of their affinity for binding to nucleic acids (*68,88,89*). Biota generally metabolize PAH in two stages. The first stage, in which potentially the most mutagenic compounds are produced, involves conversion to phenolic and other hydroxylated forms via the MFO enzyme system discussed earlier. The second stage of metabolism involves conjugation of the oxidation products to amino acids, glutathione or glucuronides in animals—all of which are highly soluble and usually excreted.

Metabolism of PAH to the more water soluble catabolites is evidently rapid enough to preclude any appreciable bioconcentration of the parent compound, judging from data on fish (*15*). For example, the higher metabolic rate for fluorene compared to anthracene leads to a reduced parent compound level in fish tissues and a higher content of polar metabolites (Fig. 7). When metabolism was inhibited by MFO inhibitor, PAH content in fat and as extractable parent compound was nearly quadrupled (*76*). Normal metabolism does not preclude the formation of certain metabolic conversion products which have been shown to accumulate in fish and crustaceans and to be retained longer after animals are removed from exposure to the parent compound (*4,90–93*). Indeed, a nonconjugated catabolite of benzo(a)pyrene metabolism has been thought to explain higher muscle retention of catabolite than parent compound in fish (*94*). Inhibition of the MFO enzyme system by piperonyl butoxide has been shown to increase retention of the parent compound and to reduce output of the metabolites in several organisms in an isolated ecosystem (*76*). It cannot be ascertained readily whether the increased accumulation of parent compounds retained in fatty tissue of the organisms pose a greater or lesser potential hazard in a food chain.

Heterocyclic Analogs of PAH

These polycyclic compounds are represented by benzothiophene, benzofuran, and acridine and other azaarenes. They are compounds in which an S, O or N atom occupies a position in one ring. Comparative data on the metabolism of heterocycles do not seem to be generally available (*1*), except for one study (*75*). In that study, a greater polarity of oxygen, nitrogen, and sulfur analogs of anthracene led to lower octanol partitioning. All compounds were thus bioconcentrated, and the metabolized fraction was very small. Among the metabolites produced, hydroxy and keto products were derived from anthracene and fluorene. *N*-Acetylated and *N*–methylated products were derived from the azaarene, carbazole. Sulfoxides and sulfones were derived from dibenzothiophene. Only the ketones were persistent, and indeed anthrone was more persistent than its parent anthracene, in the work cited. The other compounds dissipated rapidly.

Field studies suggest that the sulfur heterocycles (PASH) persist, in isopod tissues, along with their alkylated metabolites, at higher levels than the other PAH following an oil spill (*95–98*). One infers that sulfur–containing products in the ketone metabolic pathways did not further degrade in these field experiments; the differences are not simply explainable on the basis of bioconcentration.

In the case of an oxygen heterocycle, dibenzofuran, its octanol partitioning is low; it is bioconcentrated and only a small portion is metabolized (*75*). These workers also found that about 14% of the extractable dibenzofuran was metabolized to products substantially more polar. Presumably these products represented ring–hydroxylated compounds.

The azaarenes are thought to be metabolized like their PAH analogs, but, except for the study discussed above (*75*), comparative compound me-

tabolism data seem scarce. Acridines and quinolines show a substantially lower bioconcentration factor (BCF) in an exposure situation than they do when their BCFs are predicted from octanol partitioning data, and these facts are indicative of the metabolism of more polar compounds (*55*). Benzacridine metabolites (^{14}C, not otherwise identified) were retained in fish tissues and built up to levels well in excess of the parent benzo(a)acridine concentration (*57*). This happened despite the fact that the metabolites were converted to more polar forms than the benzacridine, and despite the expectation that these more polar forms would be readily excreted.

Of great importance in risk assessment are the recent findings that *N*-containing polycyclic aromatic hydrocarbons are highly mutagenic, whether azaarenes or amino-PAH.

Amino–PAH

These anilinelike compounds include aminopyrenes, aminofluorenes, and aminophenanthrenes. Among the PAH, relative carcinogenicity and mutagenicity had been fairly well established comparing single compounds, and showing that benzo-ring compounds, like benzanthracene, methylcholanthrene, and dibenzopyrene, were more potent compared to the simpler PAH (*4*). The recently studied basic compounds, like the azaarene class, have been added to the list of still more potent mutagens (*71*). Evidence is now accumulating that mutagenic activity may reside primarily with the amino derivatives of N–PAC (*7,63,64,98*). Of considerable interest is the observation that N–PAC having three or four aromatic rings are generally more active mutagens than compounds having a lesser or greater number of rings (*71*). N–PAC are significantly higher in coal conversion liquids than in petroleum.

Other Compounds

Chlorinated compounds resulting from industrial processes, rather than energy processes, have been reviewed elsewhere (*28*). These include DDT metabolites, butadienes, ethylenes, phthalic acid esters and several heavy metals contaminating Puget Sound waterways.

The metabolism of the paraffinic hydrocarbons, hexadecane, heptadecane and dotriacontane was compared with metabolism of five PAH in blue crabs (*90*). All of the hydrocarbons used in the study were metabolized at similar rates. The metabolites were mainly mono– and dihydroxy compounds and their conjugates, and the PAH included naphthalene, methylnaphthalene, fluorene, benzopyrene and methylcholanthrene.

Special Features of Metabolism

The important and very problematic feature of blocking of the mutagenic action of specific compounds in the presence of a complex mixture is a special feature of metabolism and may vary with classes of organisms. Mutagenicity remains a paramount consideration for indirect exposure of people to hydrocarbons through food chains, and PAH have been high on the list of these hydrocarbons. However, for petroleum sources, it seems unlikely that the particular distribution of PAH in petroleum is hazardous. Neoplasia in aquatic organisms has not been detectable at any environmentally realistic concentration of petroleum PAH, e.g., 1–50 ppb (*4*), and organisms living in petroleum–laden waters show significant enzymatic and ecosystem adaptations such that petroleum compounds are metabolized as carbon sources without evident ill effect (*99–101*). For nonpetroleum sources this situation may differ in important details. In coal liquids, the distribution of PAH is different; sulfur and nitrogen heterocycles are comparatively enriched; and entirely new classes of compounds, like the amino–PAH, have been identified as significant contributors to mutagenic activity (*7*). For exposure to used crankcase oil, toxic products of metabolism were demonstrated that evidently did not exist prior to MFO induction (*2*).

Organisms lower on the phylogenetic scale may emphasize different metabolic pathways in metabolizing PAH. While there do not seem to be substantive interphyletic differences among aquatic organisms (*4*) or other organisms (*77*), there are a few indications of different trends, which may not be phylum specific. Sea urchins, for example, seem to metabolize an alkyl–substituted naphthalene primarily through aromatic ring oxidations to arylsulfates (*102*) rather than by ring cleavage to diol expoxides, as predominates in fish (*15*). In algae, sorption predominates over metabolism (*102,103*). Invertebrates show rather irregular responses to MFO induction (*81*) and in crustaceans, the second–phase enzyme systems seem to be deficient for conjugating ring by–products. Thus, dihydrodiols and phenols are likely to be principal metabolites, rather than glutathione conjugates or mercapturic acids as in finfish. In mosquito larvae (Culex), snails (Physa), and the saltmarsh caterpillar (Estigmene), metabolic degradation of benzopyrene was less efficient than for fish (Gambusia) (*76*).

Efficiency was determined from data before and after metabolic initiation by using the MFO inhibitor, piperonyl butoxide. In the comparative study of the metabolism of five anthracene analogs (*75*), fish converted a far larger amount of the metabolically active fraction of each analog than did algae, snail or insect. Most of the material was retained in fat stores and not metabolized.

Zooplankters, like daphnids and copepods, evidently metabolize PAH, but their high lipid content leads to biconcentration of parent compound that almost wholly obscures residual metabolites (*104,105*). Residuals typically amounted to 6% or less.

Microbial populations show a wide range of differences in metabolism. To the extent microbial activity is thought to, or known to, influence food chain transmission of organic contaminants, discussion will be found below. An important review on metabolism of aromatic hydrocarbons by microbial organisms is also available (*106*). The information clearly shows that bacteria, fungi and algae have the enzymatic capacity to metabolize aromatic substrates ranging in size from benzene to benzo(a)pyrene. Fungi, in particular, undergo some metabolic reactions similar to those reported for mammalian enzyme systems, and they can produce metabolites known to be mutagenic, carcinogenic or acutely toxic to experimental animals. Whether these reactions occur under conditions similar to those encountered in nature is not known.

Critical Pathways

Key steps in the critical pathways whereby foodstuff becomes contaminated are: soil to plant, air to plant, and sediment to biota. Modeling exercises would pyramid several of these and other computational steps, e.g., from soil to plant to herbivore to people (*10*). I will cover only key transfer steps because the information available on overall pathways is fragmentary at best, and the modeling approach necessary to deal with metabolites is as yet undecided.

Soil to Plant

Some microorganisms can degrade certain aromatic hydrocarbons (AH) to CO_2 and water; others may only partially metabolize specific AH and/or require additional carbon sources (*107,108*). Since many of the micro-organisms are ubiquitous, their presence can be expected in soil systems as well as sediment systems. The relative numbers of specific organisms in a soil sample may also depend on substrate composition and time allowed for them to grow in (*26, 27, 109*). Simpler aromatic compounds like benzoic acid evidently can be metabolized as a sole carbon source, but it is thought that the more complex compounds will require concerted action of several bacteria that partially degrade the parent compound by a variety of metabolic pathways (*17, 27, 110*).

Plant uptake will undoubtedly vary as a complex resultant of microbial processes, release rate of the hydrocarbon, and plant transfer and metabolism. One can therefore expect substantive differences in uptake depending on soil and exposure conditions, as well as plant species. Among the few data available, carrots, spinach and lettuce were grown in soils contaminated with 3,4-benzopyrene at 100 ppb levels (*111*). Differences of PAH incorporation into the plants were tenfold, depending on the plant part, and uptake was *not* correlated with soil concentrations of benzopyrene. PCBs, by comparison, were found not to be taken up by root systems (*38*). In the PAH study cited, it is not clear what the metabolic products were, but another study using soybeans showed that anthracene first sorbed to soil was taken up, translocated and catabolized to lower MW products (*112*).

Currently, soil studies on the behavior of synfuels residuals are focusing on the fate of phenols and anilines, (*6*). These simpler ring compounds should be representative of how metabolites of the higher ring number PAH will be handled in soil systems. Alfisol soils, for example, retard the movement of organic nitrogen bases (anilines) while phenols remain relatively mobile (*113,114*). Thus, phenols are likely the first compounds to affect water quality. In contrast, the primary environmental impact of the anilines may result from plant uptake and subsequent food chain incorporation. Of the fraction that enters the plant, a portion of phenols or the anilines is metabolized. Using ^{14}C-labeled phenols, 80% or more of the ^{14}C associated with root solubles was metabolized to higher MW fractions; by contrast, using ^{14}C-aniline less than 40% of root solubles was metabolized to higher MW compounds, and the remainder persisted in tissues as parent aniline and possibly closely related compounds (*113*).

Air to Plant

Radiological experience shows that particulates of 1 μm AMAD provide the largest pathway whereby green plants take up airborne radioelements, and such contaminants are tenaciously

held by the plant leaf (*20*). This is not true of particles as large as 100 μm. Most PAH are sorbed to fine atmospheric particulates (1 μm). Recently mutagenically active hydrocarbons were found sorbed to urban air particles and absorbed to a disproportionately high degree in size class < 1.7 μm compared to size class <20 μm (*39*). Distance-dependent decreases in PAH sorbed to vegetable leaves were demonstrated in vicinity of a smelter (*115*) and near a highway (*116*). PAH are converted to more mutagenic nitroderivatives in the presence of atmospheric NO_2; the reaction is evidently catalyzed by sorption onto the airborne particulates (*41,42*).

Because they may stabilize organic pollutants against decomposition, and because of their sorption surface area and charge characteristics, airborne particles would seem to be a much more important vehicle for exposure of either plants or animals, as compared to gaseous contaminants (*9,19*). In the highway and smelter studies cited above there were also suggestions that low MW PAH might be more efficiently sorbed than high MW PAH. These inferences might well have been confounded by a particle size/sorption variable because, as is common with most monitoring studies, provision was not made for aerosol particle sizing at the sampling sites.

PCBs are thought to be taken up by plant leaves after volatilization from the soil surface, and they are not absorbed through the roots (*38*). In a field situation, or even certain growth chamber situations, wind resuspension of surface soil particulates cannot usually be ruled out for observations of this sort (*117*). Differences in how various plants handle foliar PCB; vary by two orders of magnitude (*118*). By comparison. ^{14}C-anthracene, administered by volatilization to soybean plants, was absorbed through the leaves and metabolized to lower MW products (*112*), as would be expected.

Sediment to Biota

This link in food chains to people is important simply because many complex organic compounds (and metals) are ultimately deposited in estuarine and other aquatic sediments. Re-entrainment takes place via microbial conversion, uptake from the interstitial water of sediments and ingestion of bottom particulates by some organisms used as food. In aerobic sediments, as in soils, the complete conversion of complex hydrocarbons to CO_2 and water is known to require a suite of organisms and a period of adaptive grow-in of populations not initially present in great number. During an interval of time, which may vary appreciably with temperature and other field conditions, microbial conversion is likely to be incomplete, and a wide variety of intermediate metabolites may be produced (*109*). Further research will be needed to delineate metabolites and conditions that might be important for food chain uptake.

Only recently has information been available on the distribution of sedimentary compounds that find their way into organisms directly living in the sediments. For example, a detritus feeding clam, *Macoma inquinata,* and a burrowing polychaete, *Abarenicola pacifica,* both entrained several times the sediment concentration of three PAH over a sixty day exposure (*119*). The three PAH were freed from impurities by silica gel chromatography before being sorbed to clean sediments. Tissue-bound forms of the PAH that were solvent-insoluble were considered not to be parent PAH. They amounted to 1% and 9% for chrysene and benzo(a)pyrene, respectively, and 22% for phenanthrene. It would be useful from a modeling perspective to know whether such bound forms of PAH consisted of insoluble metabolites or rather represented parent compound that had partitioned into lipid constituents. Evidently the clam was a temporary reservoir, at least, for PAH transfer to people. The polychaete would be of less concern, since an aquatic carnivore with hydrocarbon degrading capabilities would intervene in the food chain. Generally, it would seem, the principal pathway for uptake from the sediment will be via interstitial water to the filter feeder, and not directly from the sediment particle (*120,121*).

In subsequent work on New York Bight, fluoranthene, pyrene and other PAH of high MW found in the digestive gland of lobster, sharply contrasted with the predominantly lower MW compounds like napthalenes and biphenyls found in the liver of flatfish (*122*). The author considered that these differences reflected primarily habitat and feeding habits of the lobster, not differences in its metabolism compared to fish. Attention also was called to the human health hazard from high PAH content in lobster digestive gland, which is used as food.

Volatilization from spill-contaminated sediments has been thought to be a factor reducing the likelihood of reentrainment of hydrocarbons from sediment to biota (*123,124*). Later studies, in which Henry's Law coefficients were determined for two- three- and four-ring PAH, suggest that volatilization cannot significantly reduce the sedimentary concentrations of the more carcino-

genic three- and four-ring compounds (*125*). Naphthalenes and PCBs, on the other hand, may be substantially reduced in aquatic sediments by volatilization (*28,125*).

Future Direction

Research on the biologically active components of petroleum, synthetic fuel oils, and other organic products are now providing valuable generic information needed to predict the long term fate of buried waste organic constituents and the likelihood of residual chemicals entering food chains. Now that examples have become available of compound classes for which metabolites may be of greater concern than the parent compound, need exists to better characterize the subsequent behavior of key metabolites, as for example, anthrone and other ketones from PAH metabolism. The behavior of these metabolites also must be determined in plants, herbivores and other food chain organisms. In addition, the specific feeding habits and living habits of specific organisms in a food chain must be documented, because such animal behavior can bias the physiological chemistry affecting certain compounds.

Most complex mixtures, like petroleum, coal liquids and shale oil, show several unique properties unlike those exhibited by simple solutes. Of first importance is the demonstrated potentiation, as well as blocking, of mutagenic activity of specific compounds by other fractions in a complex mixture of ring-type compounds. There is clear need to characterize and better delineate the specific compounds responsible for both blocking and synergism. Biologically meaningful criteria should also be developed for supplanting the presently inadequate reference compound lists proposed for study of complex mixtures.

Because of complexity of these environmental processes, models are needed to ascertain data base needs, but current model designs are of limited usefulness and are misleading. For example, one cannot confidently predict biological risk based simply on accumulation criteria; e.g., either measured, or estimated from octanol/water partitioning. In developing a new modelling approach, attention must be paid to gradient systems, which are nonsteady-state, and means need to be devised for handling the movement of those metabolites likely to be more hazardous than the parent compound. Since compound classes differ markedly in soil persistence, metabolizability, and tissue retention following depuration, byproducts of a compound may conceivably become more toxic to organisms over time as a result of the nature and magnitude of intervening processes. Understanding the processes holds the key to the credibility of predicting health hazards from indirect exposures to organic chemicals.

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