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Contaminant BIOAVAILABILITY in Soil and Sediment

Improving risk assessment and remediation rests on better understanding bioavailability.



ioavailability refers to the extent to which humans and ecological receptors are exposed to contaminants in soil or sediment. Although long employed in toxicology and agricultural sciences, the concept of bioavailability has recently piqued the interest of the hazardous waste industry as an important consideration for deciding how much waste to clean up. The rationale is that if contaminants in soil and sediment are not bioavailable, then more contaminant mass can be left in place without creating additional risk. To date, the concept has affected cleanup goals at a small number of sites (Table 1), and enthusiasm for the idea is growing.

After two years of deliberation, a National Research Council (NRC) committee recently weighed in on using bioavailability in soil and sediment management. This article summarizes the resulting report, *Bioavailability of Contaminants in Soils and Sediments: Processes, Tools, and Applications* (1).

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TABLE 1

Select bioavailability sites

Listed below are sites where bioavailability was used to refine human health risk assessment and alter a soil cleanup goal.

Site	Contaminant	Test	Bioavailability adjustment ^a (%)	New cleanup level ^b (mg/kg)
Anaconda, Mont.	Arsenic	In vivo—monkey	18.3	250
Jasper County, Miss.	Lead	In vivo—swine	60 and 80°	800
Oak Ridge National Laboratory, Ten	n. Mercury	In vivo, in vitro, speciation	n 10	400
Palmerton, Pa.	Lead	In vivo—swine	60	650
National Zinc Co. National Priorities List Site, Bartlesville, Okla.	Lead Cadmium Arsenic	In vivo—rat, speciation In vivo—rat, speciation In vitro, speciation	40 33 25	925 100 60
Former Coal Tar Manufacturing Site Chicago, III.	, PAHs	In vivo—mouse	18	Reduced area of remediation

^aThese values are for relative bioavailability.

Regulatory issue

Bioavailability concepts have not received widespread regulatory and public acceptance, because they are sometimes perceived as "do nothing" or "do less" approaches, akin to the way natural attenuation is often viewed. Indeed, the report states that "rarely are bioavailability studies undertaken simply to improve the accuracy of a risk assessment. Rather, they are performed to justify site cleanup goals that are more financially or technically feasible and that involve leaving appreciable amounts of contaminant mass in place, while still being protective of public health and the environment." The report argues that bioavailability is inherently part of risk assessment and thus does not present a unique risk communication problem. Nonetheless, the following certain technical components should be included in any public discourse regarding bioavailability. These aspects include the factors that control contaminant bioavailability at the site, the concepts of absolute and relative bioavailability, the basis for the toxicity values, how certain models were selected for bioavailability studies, how uncertainty was handled, and how site-specific bioavailability information will be incorporated into the risk assessment.

Regulatory acceptance of the bioavailability concept for use in hazardous waste risk assessment is uneven across different regions and different types of media. The NRC committee found no legal recognition of "bioavailability" for soil cleanup, although bioavailability concepts are emerging for sediment management and have been embraced for biosolids management. A survey of U.S. EPA regions conducted by the committee revealed that acceptance and use of bioavailability in state and federal soil cleanup projects are limited at best. Although there are regional differences in the nature, types, and costs of cleanups, hesitancy to use site-specific measurements of bioavailability also reflects agency concern about increased analytical costs, public acceptance, legal challenges, and the absence of more formal national guidance. To date, there is no national policy on bioavailability, although Peter Grevatt of EPA headquarters states that "EPA is currently developing a guidance on the use of bioavailability information at contaminated sites ... and hosted a workshop in April 2003 to discuss the collection and use of site-specific bioavailability data for human health risk assessment of metals." Indeed, the report recommends EPA produce guidance that addresses what information must be included in a bioavailability assessment, its scientific validity, acceptable tools and models, and other issues.

The NRC report notes that the potential for the consideration of bioavailability to influence decisionmaking is greatest where certain chemical, environmental, and regulatory factors align. First, the contaminant whose bioavailability is under investigation must be and remain the risk driver at the site. This suggests that where several hazardous chemicals are present or where contamination has not been well characterized, time and money would be better spent on site characterization or other activities rather than on bioavailability assessment. Second, consideration of bioavailability could make a significant difference where the default assumptions made during risk assessment that affect the final cleanup goal are suspected to be inappropriate. For example, a common default assumption is that humans or ecological receptors are exposed to the total contaminant mass in soil or sediment at an affected site may not be the case and could be tested with bioavailability tools. Third, experience has shown that considering contaminant bioavailability makes sense at sites where a significant change to remedial goals is likely, such as situations that involve substantial quantities of contaminated soil or sediment. "You can achieve cost and time savings if a bioavailability assessment leads to a large amount of material [soil or sediment] being left on site or untreated, which is why the focus thus far has been expansive mining sites in the West," states committee member Michael Ruby.

Cleanup levels at these sites numerically increased because of the site-specific bioavailability adjustment, with the exception of the Palmerton, Pa., site. There are two numbers because more than one soil was analyzed. Both values were used in the risk assessment.

The report warns that bioavailability arguments should only be used to alter cleanup goals when site conditions are unlikely to change substantially over time. Actions that might make a contaminant available at a later date, such as a change in land use that creates new exposure pathways, may necessitate restarting remediation. Therefore, the report advocates a new approach to long-term monitoring—one that specifically investigates the state of the contaminant sequestered in the solid phase over time.

Defining bioavailability processes

Bioavailability has been defined differently by various disciplines, which has confounded use of the term. For this reason, the NRC report contains no explicit definition of bioavailability. Rather, it defines "bioavailability processes" as the individual physical, chemical, and biological interactions that determine the exposure of organisms to chemicals associated with soils and sediments. Figure 1 is a schematic of bioavailability processes in soil or sediment. Central to the report is that mechanistic understanding of these processes is highly variable, and that quantitative descriptive models of bioavailability processes are lacking in most cases.

The processes in Figure 1 can be thought of as barriers that must be overcome for a contaminant in soil or sediment to become bioavailable. Typically, a few steps will be most restrictive, and these are expected to control bioavailability. A refers to the physical, chemical, and biochemical phenomena that bind, unbind, expose, or solubilize a contaminant associated with soil or sediment. Contaminant-solid binding may occur by adsorption on solid surfaces, by absorption within a phaselike natural organic matter, or by precipitation, whereas contaminant release to fluids in contact with the soil or sediment occurs in response to changes in water saturation, in water and gas chemistry, and in solid surface properties. Time is an important aspect governing contaminant-solid interactions discussed in the report. Contaminants are transformed or incorporated into more stable solid phases over time, which can lead to a decrease in contaminant bioavailability-a process referred to as aging.

B involves the movement of a released contaminant to the membrane of an organism, whereas C involves the movement of contaminants still bound to the solid phase. Although not often associated with bioavailability, Processes B and C are integral fate-and-transport processes that can control an organism's overall exposure. Nonetheless, if contaminant release from the solid phase occurs internally (as in the gut lumen), fate-and-transport processes prior to uptake across a biological membrane may be limited.

D entails movement from the external environment through a physiological barrier and into a living system. Because of the enormous diversity of organisms, the actual process of contaminant uptake into a cell—or factors that may impede or facilitate uptake—varies depending on receptor type. Human uptake mechanisms include absorption across the gut wall, the skin, and the lining of the lungs. One common factor among all organisms is the presence of a cellular membrane that separates the cell interi-

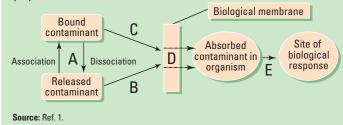
or from the external environment, through which most contaminants must pass before deleterious effects on the cell or organism occur.

E refers to paths taken by the chemical following uptake across a membrane, for example, metabolic processing or exerting a toxic effect within a particular tissue. Of particular importance is the bioaccumulation of contaminants within tissues that are often inaccessible to normal elimination mechanisms, such as metabolism and excretion. Slow release of the chemicals from these storage sites can cause protracted "exposure" within the body even when the external exposure has been reduced. In addition, bioaccumulated contaminants (e.g., polychlorinated biphenyls) may become available at some point to higher-order organisms that eat the plant or animal in which the contaminants are stored.

The NRC report's definition of "bioavailability processes" incorporates all the steps that take a chemical from being bound or isolated in soil or sediment to being absorbed into an organism (A–D in Figure 1). Although of great importance in determining the overall effect of a contaminant on an organism, E processes are not considered bioavailability processes per se because soil and sediment no longer play a role. They are included in the NRC report because many bioavailability tools measure E processes, and they are often of importance to stakeholders.

FIGURE 1 Bioavailability processes

In both soil and sediment, processes that determine exposure to contamination include release of a solid-bound contaminant (A) and subsequent transport (B), transport of bound contaminants (C), uptake across a physiological membrane (D), and incorporation into a living system (E). Note that A, B, and C can occur internal to an organism, such as in the lumen of the gut. The NRC report defines A, B, C, and D to be bioavailability processes, but not E, because soil and sediment no longer play a role.



Tools, tools, and more tools

A substantial portion of the report is devoted to evaluating the many physical, chemical, and biological tools and methods used to measure bioavailability. The tools range from physicochemical techniques, such as microscopy, to chemical extractions to various bioassays. One conclusion is that tools that further mechanistic understanding and promote predictive model development are preferred over conventional empirical approaches.

Relatively new to the assessment of bioavailability are instruments that explore the geochemical compartments that contain the contaminant, the forms of the contaminant, and interactions of the

contaminant within the solid phase. These include X-ray diffraction (XRD) and scanning electron microscopy (SEM), microscale surface mass spectrometric (microprobe two-step laser mass spectrometry [μ L²MS] and secondary ion mass spectrometry [SIMS]) and infrared spectroscopic methods, nuclear magnetic resonance (NMR), and X-ray absorption spectroscopy (XAS). Because of the sophisticated, specific nature of these instruments, they are more useful as research tools and unlikely to gain widespread use at hazardous waste sites. Nonetheless, such techniques can provide critical mechanistic information that can help interpret the results of other

bioavailability tools, such as when XAS was paired with extraction and toxicity tests to study the bioavailability of contaminated estuarine sediments in San Francisco Bay (2).

Dozens of simple, empirical extraction tests—some of which evolved from tools originally designed to measure plant uptake of nutrients—are currently used to estimate the bioavailable fraction of a contaminant pool (3–6). Many include a solid-phase sorbent, such as diffusive gradient in thin films (DGT), solid-phase microextraction (SPME), semipermeable membrane devices (SPMD), and C-18- and Tenax-containing disks or beads. For human health risk as-

TABLE 2

Ranking select bioavailability tools

Three selected bioavailability tools are ranked according to seven criteria. Tools are ranked 1 to 3, on the basis of their score in each category. Three represents the best score in a category. Definitions for abbreviations are found in the text.

Criteria	Probes for specific forms of the contaminant bound to solids (XRD, SEM, XAS, μL ² MS, SIMS, NMR)		Field survey: whole-organism bioaccumulation (Plants, invertebrates, fish, birds, mammals)
Application to the field	Some methods are hard to use on natural particles. Equipment detection limits cause problems in natural settings.	2–3 Field soils and sediments can be used but must be removed from field for test. May be able to use SPME, SPMD, and DGT in situ.	3 In situ test
Application to solid phase	3 Directly applicable to solid phase	3 Directly applicable to the solid phase or slurry	2 Integrates exposure from all influential media, including solid phase
Single versus lumped processes	3 Uniquely suited to identify mechanisms of association	Operational measure that lumps [combines] multiple processes	Whole-organism bioaccumula- tion integrates influences of several biological processes but is indicative of bio-uptake
Immediately relevant to entry into living cell	1 Requires inference about links between specific form and bio- uptake	2 Biomimetic but still an inferen- tial link to bio-uptake	Can be used to directly measure bio-uptake, but as an integrated response to influential biological and physicochemical processes
Ability to generalize	2 Will eventually be essential to generalizing about bioavailabili- ty processes	2 Reliability of generalizations about bioavailability is unproven. Work in progress.	1 Generalization possible only if data are available for a broad array of sites or situations
Relevance to regulation	1 Complicated and consequently of limited use in regulatory environment	Regulators seldom use such information for soil/sediment criteria; may be useful eventually.	3 Concentrations can be used to regulate exposure, but guidance is limited, especially for ecosystems.
Usefulness as a research tool	3 Potential to understand what controls bioavailability processes	3 Potential to measure processes important to biouptake (e.g., can obtain rates of release)	2 Mostly a tool for empirical measurements, but commonly used for research applications

sessment, in vitro extractions that mimic mammalian digestive processes are becoming common, although most have yet to be validated (7, 8).

NRC's report notes that most simple extractions, excluding in vitro extractions for human health risk assessment, cannot account for many complicated uptake mechanisms that control an organism's overall dose, such as dietary uptake or ligand complexation of contaminants. In addition, extraction procedures generally do not remove metals or organic contaminants from specific components of soils and sediments, nor can these procedures reveal the character of the solid phase to which a contaminant may be sequestered. "Because extractions are not mechanistic methods for estimating contaminant bioavailability, they are, at best, useful as screening tools in sediments," according to committee member Sam Luoma.

Bioassays, which are used to study both influential biological processes and physical and chemical processes, range from tools that measure contaminant transport across a membrane, such as assimilation efficiency (9) and isolated organ tests (10), to tools that measure responses at more complex levels of organization, such as whole organism bioaccumulation measured in feeding studies with invertebrates (11), plants (12), fish (13), birds (14), and mammals. To date, only one bioavailability feeding study has been conducted using human test subjects (15). Other tests measure even more complicated biological responses or groups of processes, such as protein and genetic biomarkers (16) and acute and sublethal toxicity tests (17). Many of the biological tools discussed in the report represent the state of the art or require additional research in order to reach their potential, especially molecular tools such as gene expression techniques (18) and reporter systems (19).

Table 2 is a highly condensed version of a table in the NRC report that ranks the strengths and weaknesses of all tool types. The criteria used to evaluate the tools are listed in the table. Tools are ranked from 1 to 3, with 3 representing the best score in a given category. No one method achieves the highest rating in all categories and none completely fail, illustrating that every tool has trade-offs. The criteria reflect the NRC committee's opinion that mechanistic approaches that determine the form and associations of a contaminant have the greatest potential for ultimately defining bioavailability and narrowing uncertainties, although they are less applicable at present.

Because more and more bioavailability tools are being developed, there can be confusion regarding which tools are best and how many to choose, especially where ecological receptors are the main concern. Common practice today consists of applying to the soil or sediment under investigation a battery of mainly operational or correlative assays that are related to contaminant bioavailability (2, 20). "We found numerous examples of this [practice], where as many as seven different tools were used at a site, sometimes with conflicting results," says committee member Barth Smets. In addition, tools intended for other purposes are sometimes misapplied to contaminant bioavailability assessment—for example, the diethyltriaminepentacetic acid soil extraction, which was

designed to predict micronutrient deficiencies in neutral to calcareous soils (21). Thus, the report advocates a "weight-of-evidence" approach to bioavailability tool selection to make near-term progress at sites. This involves initially relying on operational tools along with an intensive effort to develop tools and conceptual models based on mechanisms (22). Many operational tools, such as extractions, normalizations, and simple models, have proven ambiguous or show large uncertainties in their estimates of bioavailability when rigorously tested. They are also poorly correlated with bioavailability across ranges of environmental conditions, such as soil type. Nonetheless, there are efforts to better validate operational tools, and those that are validated should be given greater weight than those tools that are not, according to the NRC report. As more robust mechanistic methods evolve, the need for the weight-of-evidence approach is expected to diminish.

Demystifying bioavailability in risk assessment

One of the NRC committee's goals was to show how and where bioavailability processes are currently dealt with in ecological and human health risk assessment, because improving the accuracy of the risk assessment is a primary benefit of bioavailability analysis. According to the report, bioavailability is usually reflected in default values or site-specific data that are inserted into exposure equations. Although a multitude of processes can affect bioavailability (see Figure 1), a typical assessment generates only one value that ultimately adjusts the applied dose. "The net result of this single-value approach is that many bioavailability processes are hidden within risk assessment, and assumptions made about these processes are not clear. We've provided a few examples of these hidden assumptions for different exposure pathways," says committee member Richelle Allen-King.

In human health risk assessment, two operational definitions are used—absolute and relative bioavailability. Absolute bioavailability is the fraction of the applied dose that is absorbed and reaches systemic circulation. Relative bioavailability represents a comparison of absorption under two sets of conditions—for example, from a soil sample versus food—and can be greater than or less than 100%. These values are then used in exposure assessments, particularly for direct ingestion of soil or sediment and dermal contact.

Studies using animals as surrogates for humans to determine relative bioavailability—and, to a lesser extent, absolute bioavailability—for different chemical—solid combinations have been conducted at only a small number of sites. This scarcity of data has led to the extensive use of default adjustment factors for chemical absorption in human health risk assessment, mainly for dermal contact and oral ingestion of soil. In fact, Table 3, abbreviated from the report, lists default values that different federal and state regulatory agencies regard as acceptable for individual compounds and groups of compounds. According to the NRC report, the most prominent default is that relative bioavailability is assumed to be 100% unless there is compelling evidence to the contrary and a

scientifically defensible adjustment factor can be derived. This is usually a conservative assumption, states the report, because most toxicity tests intentionally use forms of a chemical that are readily absorbed.

Nonetheless, the NRC report warns that "default values used to represent certain bioavailability processes in risk assessment may not be protective and appropriate for all circumstances." For example, although the national default value for oral ingestion of lead in soil corresponds to a relative bioavailability of 60% (23), actual site-specific measurements of lead relative bioavailability can vary significantly from this value, as shown in Figure 2. Thus, the report encourages replacing default values with site-specific measurements—something that has occurred for a small subset of human health risk assessments across the country (see Table 1) and which has yet to be acknowledged in laws or regulations for hazardous waste cleanup at the federal or state level.

Bioavailability processes are more frequently accounted for in ecological risk assessments, although they have not been labeled as "bioavailability assessments or adjustments" per se. Compared with human health risk assessment, ecological risk assessment is more complex because of the many species, physiologies, and physicochemical processes to consider. "We focused on bioavailability for direct contact of invertebrates and the wildlife exposure pathway because they frequently drive ecological risk assessment," says committee member Charles Menzie. For the direct contact pathway, relatively simple techniques have been developed that predict the partitioning of metals and organics between different

phases—solid, aqueous, or within an organism—with the latter two representing the bioavailable fraction. Two popular partitioning techniques—acid volatile sulfide and the biota–soil/sediment-accumulation factor—are comprehensively reviewed in the report. "Both techniques have substantial uncertainties, and, at best, may capture only the crudest influences," according to the report.

Relating bioavailability and remediation

Limitations in our understanding of bioavailability processes have important ramifications for site management, argues the NRC report. In particular, there are treatment remedies that rely heavily on increasing or decreasing bioavailability. Without a better understanding of bioavailability processes, it is difficult to know if such treatments are effective.

Treatment technologies reported to "decrease bioavailability" generally impede transfer of a contaminant from soil or sediment to a living organism. A prominent example, particularly for polycyclic aromatic hydrocarbons, is biostabilization—that is, using bioremediation to reduce contaminant mobility and toxicity of contaminated soils and sediments. Sediment capping-for example, putting clean sand on top of contaminated sediments-accomplishes similar ends by reducing the access of bottom-dwelling organisms to the contaminant pool and by increasing mass transfer distance. Another option, particularly for metal-contaminated sites, is vitrification or solidification, which decreases contaminant mobility by increasing mass transfer resistance out of the solid. Finally, chemical alteration to decrease bioavail-

TABLE 3

Example default values for bioavailability

These examples of default values are used to adjust exposures to account for reduced bioavailability of compounds in soil. References are bracketed. (OC is organic carbon in soil; SVOCs represent semivolatile organic compounds.)

Chemical	Dermal absorption factor ^a	Oral relative absorption factor ^b
Benzene	0.08 [<i>24</i>], 0.0005 [<i>25</i>]	1.0 [<i>24</i>]
Naphthalene	0.1 [<i>24</i>], 0.1 [<i>25</i>]	1.0 [<i>24</i>]
Benzo[a]pyrene	0.18 [<i>24</i>], 0.1 [<i>25</i>]	0.91 [24]
Lindane	0.04 [26]	
DDT	0.03 [26, 27]	
PCB Aroclors 1254 and 1242	0.14 [<i>26, 27</i>]	
Dioxins	0.03 or 0.001 if OC >10% [26]	
Polycyclic aromatic	0.15 [<i>29</i>], 0.05 [<i>28</i>], 0.13 [<i>26</i>];	
hydrocarbons	(0.1 for SVOCs [26, 30])	
Volatile organic compounds	0.25 [<i>30</i>]	
Arsenic	0.03 [26, 27]	
Cadmium	0.1 [<i>27</i>], 0.001 [<i>26</i>]	
Lead		0.3 [<i>23, 31</i>], 0.12 [<i>32</i>] ^c
	in addition the desired with The term "absorbed" in some	1 1 2 1 1

"The absolute bioavailability of the compound in soil via the dermal route. The term "absorption" is commonly used in human health risk assessment literature when referring to bioavailability.

^bThe relative bioavailability of the compound (i.e., in soil versus in the medium used in the toxicity study).

^eValues for lead are absolute bioavailability.

Source: Data from Refs. 24-33.

ability includes converting compounds to less soluble forms or redox states via amendment. For example, phosphorus, iron, and biosolids are common amendments for stabilizing lead in soil.

Other technologies attempt to increase pollutant removal or destruction by enhancing a contaminant's bioavailability. These technologies increase mass transfer from the sorbed phase via physical or chemical means. Examples of the former include grinding or mixing to decrease diffusional paths and increasing temperature of the solid phase to increase mass transfer rates. Chemical means include using surfactants, cosolvents, or chelating agents to increase mass transfer by enhancing the solubility of hydrophobic organic compounds or metals or by mediating changes to the solid-phase structure or the bulk aqueous phase.

In addition to these deliberate actions, the NRC report cautions site managers to be cognizant of treatment technologies that may unintentionally affect bioavailability. Especially for sediment dredging and new technologies that have yet to be fully tested like phytoremediation, there may be unanticipated side effects that result in undesirable changes in bioavailability to certain receptors.

Moving forward

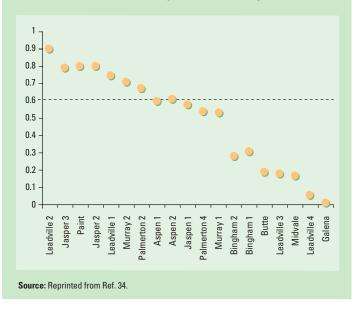
More explicit consideration of bioavailability processes in hazardous waste cleanup could refine risk assessment and, in some cases, alter the amount of contaminated material treated. In addition, bioavailability assessment can help better prioritize cleanup actions. For example, consider sites where total contaminant concentration increases in order from Site 1 to 5 but contaminant bioavailability (as measured by an unspecified method) decreases in order from Site 1 to 5. Although Site 5 has the highest total contaminant levels, it could have the lowest effective contaminant concentration depending on its limited bioavailability. Thus, in addition to refining the volume of soil or sediment requiring remediation, it is also conceptually possible to reverse the order of importance for dealing with sites if the bioavailable chemical concentration rather than the total chemical concentration is considered.

Given the potential benefits of more explicit bioavailability assessment, what can be done to overcome the resistance of regulators in allowing site-specific measurements of bioavailability to replace default assumptions? The report proposes an adaptive management approach so that bioavailability processes will be more widely considered in riskbased management of contaminated soils and sediments. This would involve pilot studies with different tools and models, and then using the results to develop a common systematic approach for how and when to incorporate bioavailability concepts into regulations in a consistent manner. An adaptive management example relevant to bioavailability is the approach recently recommended by the EPA for determining the efficacy of dredging PCB-contaminated sediment from the Hudson River. The plan involves evaluating risks over time and adjusting cleanup plans as performance monitoring data are analyzed.

FIGURE 2

Relative bioavailability of lead in soil

The graph presents relative bioavailability results from swine feeding studies using 17 field soils contaminated with lead from Leadville, Colo.; Jasper, Mont.; Murray Smelter, Utah; Palmertown, Penn.; Aspen, Colo.; Bingham Creek, Utah; Butte, Mont.; and Midvale, Utah, and two laboratory-prepared soils (paint and the mineral galena). The dashed line represents the 60% relative bioavailability value, which is used to set the EPA's current national default value for absolute bioavailability of lead in soil. The values for Leadville are highly variable because both mining wastes and smelter emissions impact soils at this large site.



Most information on contaminant bioavailability—particularly for human health risk assessments—comes from industry-funded studies at specific sites. "For a lot of different reasons, these studies are usually not conducted in a way that advances mechanistic understanding of bioavailability processes," says committee member Steve Roberts. The report notes that unless a greater commitment is made to fund bioavailability studies from a research- rather than industry-driven perspective, progress in explicitly incorporating bioavailability into human health and ecological risk assessments will be slow.

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