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**EVALUATING ECOLOGICAL RISK TO INVERTEBRATE RECEPTORS  
FROM PAHS IN SEDIMENTS AT HAZARDOUS WASTE SITES**

by

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1     **INTRODUCTION**

2             In March 2004, ORD’s Ecological Risk Assessment Support Center (ERASC) received a  
3     request from the Ecological Risk Assessment Forum (ERAF) relating to the evaluation of  
4     ecological risk to vertebrate and benthic invertebrate receptors from polycyclic aromatic  
5     hydrocarbon compounds (PAHs) in sediment at hazardous waste sites. This paper only addresses  
6     risks to benthic invertebrates because reaching a consensus scientific position on vertebrate risk  
7     issues is a longer-term prospect. Benthic invertebrates are an important component of the biotic  
8     integrity of the nation’s waters. The *PAHs* addressed in this paper are composed of carbon and  
9     hydrogen and do not include any heterocyclic atoms like oxygen, sulfur or nitrogen, or functional  
10    groups such as nitro or hydroxyl.

11            Due to the use of fossil fuels in industrialized societies and subsequent transport via  
12    atmospheric and aquatic pathways, PAHs are among the most widely distributed organic  
13    pollutants. Furthermore, because of their presence in petrochemical substances ranging from  
14    petroleum to creosote, they are found in concentrations of parts per million (ppm) in heavily  
15    industrialized sites, while in areas remote from human activity they occur in parts per trillion  
16    (ppt).

17            PAHs in the environment are known to originate from two sources: petrogenic and  
18    pyrogenic. Petrogenic PAHs originate from petroleum sources including different types of oils,  
19    coals and organic shales. Their introduction to the environment is frequently through spillage of  
20    oils during transport. Pyrogenic PAHs are produced when fossil fuels are oxidized during  
21    combustion. They are, therefore, released into the environment via the atmosphere, often  
22    associated with different forms of soot or black carbon. Eventually, these PAHs are removed

1 from the air phase through association with aerosols which then settle into terrestrial and aquatic  
2 environments.

3 There are two basic types of PAHs: parent and alkylated. These classifications involve  
4 the chemical structure of PAHs. Parent PAHs consist of only benzene rings fused together.  
5 Conversely, alkylated PAHs have various levels of alkyl substitutions added to the fused ring  
6 structure. Because of the different sources and types of PAHs, the pyrogenic parent PAHs are  
7 ubiquitous while petrogenic alkylated PAHs are more likely to be found associated with point  
8 sources like oil spills.

9 The prevalent mechanism of PAH toxicity to invertebrates is narcosis, which results in  
10 the degradation of cell membranes. This degradation can result in mild toxic effects or mortality  
11 depending upon the exposure. Some PAHs also demonstrate photoactivated toxicity. This form  
12 of toxicity can cause mortality at very low concentrations of PAHs but requires direct exposure  
13 of organisms to ultraviolet (UV) radiation in sunlight. Further, water strongly attenuates UV  
14 radiation; thus, relatively shallow overlying water will protect benthic organisms from adverse  
15 effects. The UV radiation causes the chemical bonds in the PAHs to excite and form high energy  
16 radicals, which, for a very brief time period, oxidize the tissue of exposed organisms.

17 Carcinogenicity and teratogenicity have also been reported to occur due to exposure to certain  
18 PAHs (e.g., benzo(a)pyrene), but there are limited data with regard to benthic invertebrates. In  
19 general, unless conditions result in elevated UV levels, narcosis is the most common mode of  
20 action of concern with PAHs in sediments.

21 Each of the above characteristics results in factors contributing to the nature of the PAH  
22 exposure and kinds of PAH toxic effects. In this white paper, equilibrium partitioning (EqP) is  
23 recommended for use in predicting PAH exposure concentrations, and narcosis theory is applied

1 to determine whether or not sufficient PAHs are present to cause adverse effects. A selection of  
2 PAHs defined as “total PAH” is also provided as well as an analytical method for measuring  
3 PAHs. This white paper summarizes an approach for evaluating ecological risk to benthic  
4 invertebrate receptors from PAHs in sediments at hazardous waste sites. This approach is based  
5 heavily upon the recently published EqP Sediment Benchmark (ESB) for PAH mixtures  
6 document prepared by the U.S. EPA (2003). Consequently, this white paper should be used in  
7 conjunction with U.S. EPA (2003).

## 8 **STATE OF PRACTICE**

### 9 **Invertebrate Risk Assessment**

10 A brief and limited survey of project managers, scientists and risk assessors at sites  
11 around the country, including the Pine Street Bridge Canal, Hocomonco and Beard Maguire sites  
12 in Massachusetts (Region 1), the Ashland site in Wisconsin (Region 5) and Lower Duwamish in  
13 Washington State (Region 10), indicated several characteristics of how PAH risk to invertebrates  
14 is assessed currently at contaminated sediment sites. First, there is no “standard state of practice”  
15 per se, rather, assessments are addressed on a site-by-site basis. Secondly, because of the  
16 metabolism of PAHs by many organisms at various levels of the food web, there is no clear  
17 relationship between body burdens of PAHs and effects, and hence tissue residues are seldom  
18 used as measures of exposure. Thirdly, as a consequence of PAH metabolism, exposure and  
19 effects measurements are most often assessed in the benthos, where acute and sublethal toxicity  
20 may be observed. Specifically, sediment or interstitial (pore) water measures of PAHs are used  
21 to quantify exposure while toxicity to benthic organisms is applied as a measure of effects. In  
22 some instances, benthic community composition and condition are used to assess effects.  
23 Sediment quality guidelines including empirical (Long et al., 1995; Field et al., 2002) and

1 consensus (Swartz, 1999; MacDonald et al., 2000) approaches as well as the mechanistic ESBs  
2 (U.S. EPA, 2003, 2005) are also used as complementary and predictive tools for assigning risk.  
3 In a few rare cases, photo-enhanced toxicity caused by PAHs has also been used to assess risk.

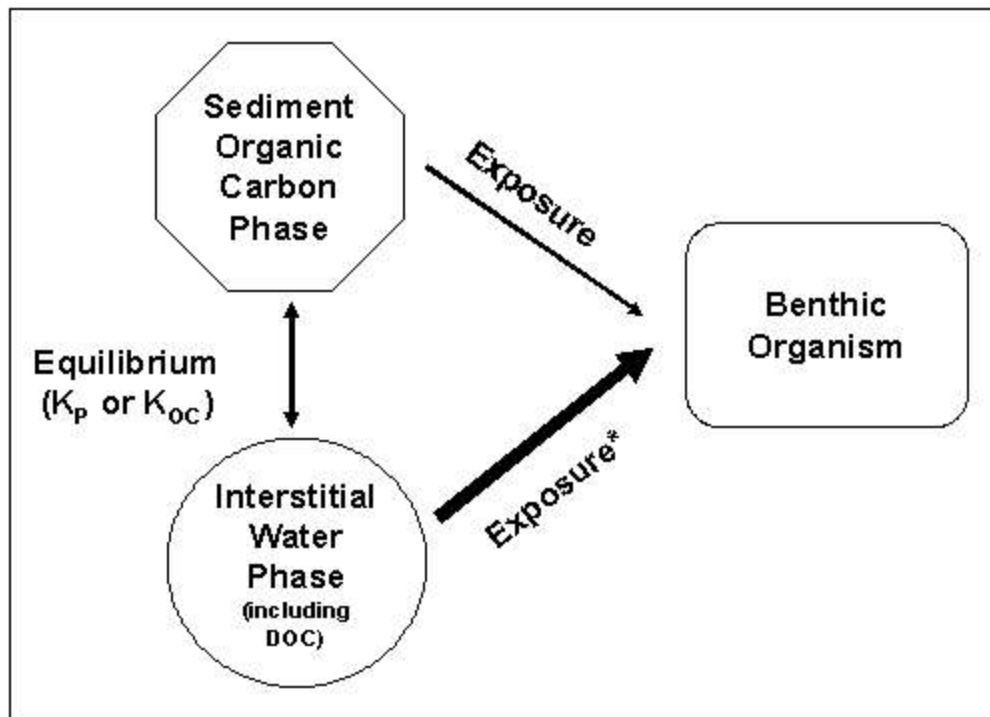
## 4 **OVERVIEW OF PAH EXPOSURE TO INVERTEBRATES**

### 5 **Use of Equilibrium Partitioning (EqP) to Predict Exposure**

6 To determine the exposure invertebrates experience in contaminated sediments it is  
7 necessary to measure or predict the concentrations of bioavailable PAHs. For hydrophobic  
8 organic contaminants like PAHs, under equilibrium conditions, the interstitial water  
9 concentration of PAH is the most accurate indicator of the bioavailable exposure concentration.  
10 The interstitial water concentration can be measured empirically using several methods (U.S.  
11 EPA, 2001). However, the results may be affected by manipulation of the sediment and  
12 interstitial water, and the methods may be logistically impractical and expensive. Measurement  
13 of the interstitial water concentration of PAHs has the additional challenge of assessing the effect  
14 of dissolved organic carbon (DOC) on bioavailability. The presence of DOC has been shown to  
15 reduce PAH bioavailability.

16 An alternative approach for determining exposure is to *predict* PAH interstitial water  
17 concentrations. The use of EqP is recommended for making such predictions. In a sediment  
18 system, the predominant phases involved in EqP include the sediment organic carbon and  
19 dissolved phase (i.e., interstitial water) (see Figure 1). Based on EqP, if the sediment  
20 concentration of PAH and concentration of sediment organic carbon ( $f_{OC}$ ) are known, the  
21 interstitial water concentration of PAH can be predicted. As discussed above, because the  
22 interstitial water concentration of PAH is the primary exposure concentration, knowing this  
23 concentration allows for an assessment of potential risk to benthic invertebrates.





**FIGURE 1. Diagram of Important Sediment Phases Affecting the Bioavailability of PAHs in Sediments**

\*The larger arrow indicating exposure between “Interstitial Water Phase” and “Benthic Organism” signifies this is a dominant exposure route.

### **EqP Model Assumptions**

Use of EqP to predict exposure concentrations comes with several assumptions: (1) the environmental system and phases therein are at or approximating equilibrium, (2) interstitial water is a good measure of bioavailable contaminant and (3) the sediment organic carbon is the primary partitioning phase for the contaminant. Recent studies demonstrate that other forms of carbon, specifically black carbon present in soots, fly ash and chars, can very strongly associate with PAHs and alter their geochemical behavior as compared to “regular” organic carbon (e.g., Accardi-Dey and Gschwend, 2002). If black carbon is suspected to be present in the sediments of interest, a site-specific prediction or measure of PAH bioavailability is necessary (Section 6.8 of U.S. EPA, 2003 discusses black carbon in detail).

## 1     **Definition of Total PAHs and Recommended Analytical Method**

2             The term *total*, when discussing PAHs is misleading. There are tens of thousands of  
3 possible PAH structures ranging from the smallest PAH naphthalene to the largest forms like  
4 coronene. Alkylation, especially in petrogenic PAHs, contributes several thousand or more  
5 varying structures of PAHs. Current technology does not allow for the direct analytical  
6 measurement of all these PAHs. Early methods for measuring PAHs focused on the 13 priority  
7 pollutant PAHs identified by the U.S. EPA. Since the mid-1980s, for NOAA’s National Status  
8 and Trends Program, 23 PAHs are routinely analyzed. Currently, the U.S. EPA’s Ecological  
9 Monitoring and Assessment Program (EMAP) measures up to 34 PAHs. Also, the U.S. EPA  
10 PAH mixtures benchmark document (U.S. EPA, 2003) recommends these 34 PAHs be analyzed  
11 when assessing the risk represented by PAHs in contaminated sediments. For the purposes of  
12 this white paper, these 34 PAHs are recommended for analysis in order to capture PAHs  
13 constituting an operational definition of “total PAHs.”

14             The 34 PAHs are listed in Table 1. Although this list is far from comprehensive, it does  
15 incorporate many of the most common parent PAHs and many alkylated PAHs frequently found  
16 in PAH mixtures. Often, a major limitation in the analysis of PAHs is the availability of  
17 standards, especially for the alkylated PAHs. As methods improve for measuring PAHs and  
18 standards become available, the list presented in Table 1 may expand to include more PAH  
19 molecules. Further, as the list of PAHs increases in number of analytes, uncertainty in method  
20 predictions will decrease. The level of uncertainty is likely to never be negligible but will decline  
21 as the most common PAHs are included in the analysis. For example, predictions made using the  
22 list of 34 PAHs will have less uncertainty than estimates using only 13 PAHs. An analytical

1 method for performing the analysis of the 34 PAHs defined above is provided in Lauenstein and  
2 Cantillo (1998).

### 3 **OVERVIEW OF PAH EFFECTS TO INVERTEBRATES**

#### 4 **Use of Narcosis Model to Predict Effects**

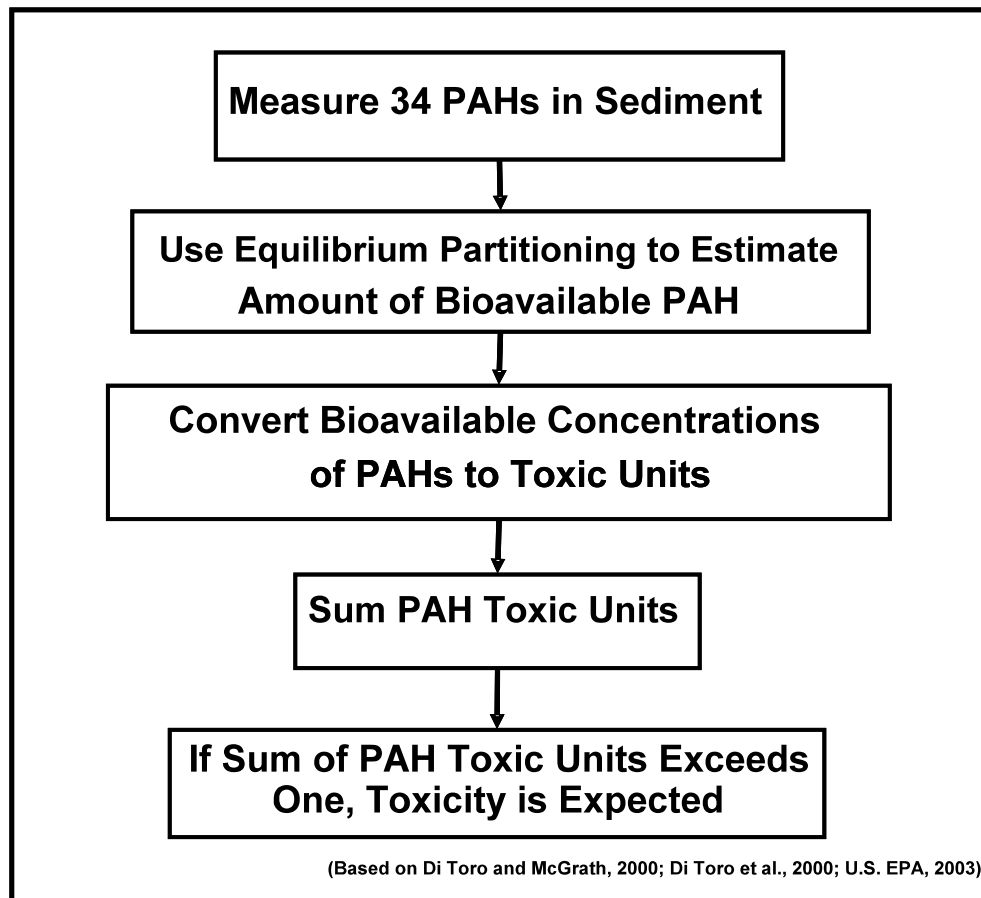
5 As discussed above, the principal form of toxicity elicited by PAHs to benthic  
6 invertebrates is narcosis. Narcotic toxicants frequently demonstrate additive toxicity; that is, the  
7 effects of narcotic toxicants can be added together to summarize the total amount of toxicity  
8 present in a mixture of such chemicals (as occurs in sediments). Figure 2 illustrates the approach  
9 used in U.S. EPA (2003) and discussed in Di Toro and McGrath (2000), Di Toro et al. (2000) and  
10 Mount et al. (2003) for predicting toxicity to benthic organisms caused by PAHs.

11 Using contaminated site sediment data, including PAH concentrations and sediment  
12 organic carbon content, EqP is used to predict the bioavailable concentrations of the 34 PAHs. As  
13 discussed in U.S. EPA (2003), the bioavailable concentration of each PAH is then converted to  
14 toxic units based on narcosis theory. The effects endpoint used to calculate toxic units in U.S.  
15 EPA (2003) are the PAH final chronic values (FCVs). The FCVs for over 60 PAHs, including the  
16 34 PAHs discussed above in Table 1, are reported in Table 3-4 of U.S. EPA (2003). These toxic  
17 units are summed together and an estimate of whether or not toxicity is expected can be derived.  
18 For example, if the sum of the toxic units exceeds a value of 1, toxicity to benthic invertebrates is  
19 expected to occur.

**TABLE 1. List of PAHs Recommended for Analytical Measurement to Quantify “Total PAHs” (from U.S. EPA, 2003)**

PAH	CAS*	Molecular Weight (µg/mol)
Naphthalene	91203	128.17
C1-Naphthalenes	-	142.20
Acenaphthylene	208968	152.2
Acenaphthene	83329	154.21
C2-Naphthalenes	-	156.23
Fluorene	86737	166.22
C3-Naphthalenes	-	170.25
Anthracene	120127	178.12
Phenanthrene	85018	178.23
C1-Fluorenes	-	180.25
C4-Naphthalenes	-	184.28
C1-Phenanthrene/anthracenes	-	192.26
C2-Fluorenes	-	194.27
Pyrene	129000	202.26
Fluoranthene	206440	202.26
C2-Phenanthrene/anthracenes	-	206.29
C3-Fluorenes	-	208.30
C1-Pyrene/fluoranthenes	-	216.29
C3-Phenanthrene/anthracenes	-	220.32
Benz(a)anthracene	56553	228.29
Chrysene	218019	228.29
C4-Phenanthrenes/anthracenes	-	234.23
C1-Benzanthracene/chrysenes	-	242.32
Benzo(a)pyrene	50328	252.31
Perylene	198550	252.31
Benzo(e)pyrene	192972	252.32
Benzo(b)fluoranthene	205992	252.32
Benzo(k)fluoranthene	207089	252.32
C2-Benzanthracene/chrysenes	-	256.23
Benzo(ghi)perylene	191242	276.23
C3-Benzanthracene/chrysenes	-	270.36
Indeno(1,2,3-cd)pyrene	193395	276.23
Dibenz(a,h)anthracene	53703	278.35
C4-Benzanthracene/chrysenes	-	284.38

\* For C# PAHs CAS is not available.



**FIGURE 2. Simplified Flowchart of Approach for Predicting Toxicity of PAHs to Benthic Organisms**

**Examples of EqP Approach in Use**

A simple example of this approach is provided below. For simplicity, three PAHs are addressed in this example rather than the recommended 34. All the information needed to work with this example, except sediment concentrations and sediment organic carbon (i.e., site data), is available in U.S. EPA (2003). As shown in Table 2, concentrations of the PAHs anthracene, fluoranthene and chrysene range from 3328 to 51,896  $\mu\text{g}/\text{kg}$  in a sediment with an organic carbon content of 0.0202  $\text{g}_{\text{OC}}/\text{g}$ . Dividing the PAH concentration by the sediment organic carbon (and again dividing by 1000 to account for differences in units) results in the organic carbon normalized PAH concentration ( $C_{\text{OC,PAH}_i}$ ). This value is a more realistic indicator of the concentration of

1 bioavailable PAHs. Dividing the  $C_{OC,PAHi}$  by an organic carbon normalized toxicity value  
 2 ( $C_{OC,PAHi,FCVi}$ ) generates toxic units for each PAH. For this example, and as used in U.S. EPA  
 3 (2003), PAH FCVs are applied to generate sediment toxicity values. These values,  $C_{OC,PAHi,FCVi}$  (in  
 4  $\mu\text{g}/\text{g}_{OC}$ ), for individual PAHs are calculated by multiplying the PAH specific FCV (in  $\mu\text{g}/\text{L}$ ) by the  
 5  $K_{oc}$  for that PAH (and again dividing by 1000 to account for differences in units); they are also  
 6 reported in Table 3-4 of U.S. EPA (2003). As noted above, if the sum of the toxic units exceeds  
 7 1.0, there is an elevated likelihood that toxicity to benthic organisms will occur. In the example  
 8 above, because of the high concentrations of fluoranthene and chrysene in the sediments, the sum  
 9 of the toxic units easily exceeds 1.0 with a value of 5.17. These sediments are predicted to exhibit  
 10 chronic toxicity from PAHs. The same basic process is used when considering all the other PAHs.

11  
 12 **TABLE 2. Example Calculation of Toxic Units Associated with a Sediment**  
 13 **Contaminated with Three PAHs**  
 14

15 PAH	Concentration ( $\mu\text{g}/\text{kg}$ )	Sediment Organic Carbon ( $\text{g}_{OC}/\text{g}$ )	$C_{OC,PAHi}$ ( $\mu\text{g}/\text{g}_{OC}$ )	$C_{OC,PAHi,FCVi}$ ( $\mu\text{g}/\text{g}_{OC}$ )	Toxic Units
16 Anthracene	3,328	0.0202	164.8	594	0.28
17 Fluoranthene	51,896	0.0202	2569	707	3.63
18 Chrysene	21,453	0.0202	1062	844	1.26
					19 $\Sigma = 5.17$

20  
 21 More complex examples of the use of this type of approach can be found in the scientific  
 22 literature (e.g., Swartz et al., 1995; Di Toro and McGrath, 2000; Di Toro et al., 2000; Ozretich et  
 23 al., 2000). In their study, Swartz et al. (1995) evaluate an early version of the approach, in which  
 24 toxic units of 13 PAHs based on sediment concentrations were used to successfully predict  
 25 observed sediment toxicity. In an extension of Swartz et al. (1995), Ozretich et al. (2000) included

1 33 PAHs in his evaluation of this type of approach using contaminated sediments from Elliot Bay,  
2 Washington. In that evaluation, the approach was generally successful in predicting observed  
3 sediment toxicity (Ozretich et al., 2000). Further, Di Toro and McGrath (2000), Di Toro et al.  
4 (2000) and Mount et al. (2003) describe in great detail the technical basis for the approach,  
5 discussing its performance in comparison to the results of toxicity testing and EMAP benthic  
6 analyses (Di Toro and McGrath, 2000).

7 Finally, in U.S. EPA (2003) three ‘real-life’-like examples are provided and discussed in  
8 detail in Section 6.3. To enhance the realism of the examples, the authors include scenarios where  
9 only 13 PAHs were measured as well as cases in which all 34 PAHs were quantified.

#### 10 **PAH Datasets**

11 Frequently, especially in the case of older data sets, fewer than the 34 recommended PAHs  
12 were measured. Under some conditions, the toxic units contribution of PAHs not measured can be  
13 predicted using uncertainty factors (Section 6 in U.S. EPA, 2003). In principle, the uncertainty  
14 factor serves as a multiplier to convert the toxic units associated with 13 or 23 measured PAHs to  
15 the toxic units of the desired 34 PAHs based on a selected confidence level (e.g., 95%). However,  
16 due to the unique distribution of PAHs in contaminated sediments resulting from their original  
17 source(s), uncertainty factors tend to be very site-specific. Consequently, the uncertainty factors in  
18 U.S. EPA (2003) should only be used to provide a very general estimate of the toxic units  
19 associated with 34 PAHs. Further, if only 13 or 23 PAHs have been measured in the contaminated  
20 sediments of interest, the development of site-specific uncertainty factors using a subset of  
21 sediments from the site is highly recommended. Site-specific uncertainty factors would provide a  
22 cost-effective way to reduce the variability around the predicted toxic units at the contaminated  
23 site.

1     **Model Assumptions**

2             The approach described above for predicting risk to benthic invertebrates from sediment  
3     PAHs also requires several assumptions including the following: (1) benthic invertebrates do not  
4     appreciably metabolize PAHs, (2) the PAHs used to make predictions of toxicity are composed of  
5     carbon and hydrogen and do not include any heterocyclic atoms like oxygen, sulfur or nitrogen, or  
6     functional groups such as nitro or hydroxyl, and (3) the invertebrates for which risk is being  
7     predicted are coupled to the benthic environment; that is, they are exposed to toxic chemicals  
8     primarily via the sediment. See Di Toro and McGrath (2000), Di Toro et al. (2000), Mount et al.  
9     (2003) and U.S. EPA (2003) for further discussion of these assumptions.

10            It is worth noting that a sum of toxic units greater than 1 can occur without the occurrence  
11    of significant benthic organism toxicity. This may happen if another sediment phase, like the black  
12    carbon discussed earlier, is reducing PAH bioavailability. Further, sediment toxicity to benthic  
13    organisms can occur if the sum of toxic units is less than 1, but this will most likely be due to the  
14    presence of other toxicants including, possibly, unanalyzed PAHs.

15     **SUMMARY**

16            This white paper provides an overview of an approach for assessing risk to invertebrate  
17    receptors resulting from exposure to PAHs in contaminated sediments at hazardous waste sites.  
18    PAHs are possibly the most widely distributed of anthropogenic organic pollutants. The approach  
19    is based on the procedures described in U.S. EPA (2003) and involves the use of EqP to determine  
20    exposure/bioavailability and narcosis theory to estimate sublethal toxicity of PAHs to benthic  
21    invertebrates. The white paper also provides examples of how to use this approach with analytical  
22    data resulting from the analysis of contaminated sediments. The approach, particularly when used  
23    with other contaminated sediment assessment methods, offers risk assessors a useful tool for



1 assessing the risk of PAHs to benthic invertebrates at hazardous waste sites. Whenever possible,  
2 assessments of sediments are improved when multiple lines of evidence are used (Adams et al.,  
3 2005).

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