# Multimedia Chemical Fate Model for Environmental Dredging

Fabian F. Sanchez<sup>1</sup>; Louis J. Thibodeaux<sup>2</sup>; Kalliat T. Valsaraj<sup>3</sup>; and Danny D. Reible<sup>4</sup>

**Abstract:** Dredging the bed-sediment of rivers, lakes, estuaries, and harbors is a process of remediating contaminated sites. Whether used alone or in conjunction with in situ capping or natural recovery, its application results in the loss of contaminants to the surroundings. A steady-state multimedia box model has been developed consisting of a dredge element that initiates chemical release in a flowing stream and an algorithm for residue transport to air, water, and sediment, both with and without silt curtain containment. The model quantifies these losses and, commencing with the original in-place contaminant mass, determines the quantity targeted for dredging and the quantity "delivered to shore." The latter is a measure of the efficiency of the dredging operation. The model was developed from experience gained studying the effectiveness of environmental dredging at three sites: Bayou Bonfouca (Slidell, La.), Grasse River (Massena, N.Y.), and Manistique Harbor (Manistique, Mich.). This paper describes the multimedia model theory and algorithm development and demonstrates its use in the application to the Bayou Bonfouca Superfund site, which was dredged for removal of creosote waste containing 16 polyaromatic hydrocarbons.

## **DOI:** 10.1061/(ASCE)1090-025X(2002)6:2(120)

CE Database keywords: Multimedia; Dredging; Models; Chemicals; Sediment; Contamination; Environmental impacts.

## Introduction

Toxic and hazardous materials exist in the sediment on the bottom of rivers, lakes, and estuaries in the United States. This accumulation of contaminants in surface waters represents a long-term source of contamination to aquatic life. Direct human contact with contaminated sediments is also a concern if they are present in a shallow water environment used for recreational purposes.

For contaminated sediments, remediation technologies can be grouped into four categories: interim control, in situ management, sediment removal and transportation, and ex situ management (NRC 1998). Interim control is a means of reducing risk of human exposure by implementing preventive measures (warning signs, fencing, swimming and fishing advisories, and so on). In situ management deals with the contaminated sediment on-site, where the technologies are for containment and treatment. A form of containment is capping, which is the process of placing a layer of clean sediment such as sand over the contaminated bed. Another form of in situ management is monitored natural attenuation, which entails allowing natural processes to reduce the contaminant concentration and/or bioavailability.

Note. Discussion open until September 1, 2002. Separate discussions must be submitted for individual papers. To extend the closing date by one month, a written request must be filed with the ASCE Managing Editor. The manuscript for this paper was submitted for review and possible publication on February 9, 2001; approved on December 26, 2001. This paper is part of the *Practice Periodical of Hazardous, Toxic, and Radioactive Waste Management*, Vol. 6, No. 2, April 1, 2002. ©ASCE, ISSN 1090-025X/2002/2-120-128/\$8.00+\$.50 per page.

The two removal technologies involve relocation and handling of the contaminated sediment. Sediment removal is accomplished using dredges or earth-moving equipment. Ex situ management is accomplished by placing the dredged material in landfills, confined disposal facilities, or confined aquatic disposal facilities on or near the contaminated sites. Alternative and more aggressive ex situ treatment technologies such as incineration are site specific, and their implementation depends on the cost and environmental friendliness of the operation and other factors.

Of all the remediation technologies available, the U.S. Environmental Protection Agency (USEPA) mandates that most of the sites designated for cleanup consider dredging. In fact, "dredging has become the default remedy for contaminated sediments" (GEC 2000). Environmental dredging operations remove sediments with some known contamination as effectively as possible. An effective method would include complete removal of the desired sediment with as little environmental risk and consequence as possible (Hayes 1992).

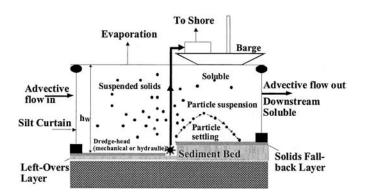
Anticipating extensive use of environmental dredging, it is important to be able to determine the effectiveness of the operation. The General Electric Company (GEC) recently compiled a report on its effectiveness in controlling risks. The report reviewed major sediment remediation projects in the United States, their objectives, the technologies being employed, and the capabilities and limitations of those technologies (GEC 2000). The primary conclusions drawn from the 54 sites reviewed were (1) environmental dredging has not reduced surface sediment concentrations to acceptable levels, being higher in some cases (that is, Manistique Harbor, Mich.); (2) dredging has not been shown to lead to quantifiable reductions in fish contaminant levels; (3) dredging projects are costly and take a long time to complete; (4) dredging resuspends and releases contaminants into the water column; and (5) dredging technology and experience in large rivers are limited. From a chemodynamic standpoint, the main disadvantage to dredging is the resuspension of sediment in the water column. Sediment resuspension leads to contaminant releases in the water column and other environmental media (air, soil, and sediment).

<sup>&</sup>lt;sup>1</sup>Gordon A. and Mary Cain Dept. of Chemical Engineering, Louisiana State Univ., Baton Rouge, LA 70803.

<sup>&</sup>lt;sup>2</sup>Gordon A. and Mary Cain Dept. of Chemical Engineering and South/ Southwest Hazardous Substance Research Center, Louisiana State Univ., Baton Rouge, LA 70803.

<sup>&</sup>lt;sup>3</sup>Gordon A. and Mary Cain Dept. of Chemical Engineering, Louisiana State Univ., Baton Rouge, LA 70803.

<sup>&</sup>lt;sup>4</sup>Gordon A. and Mary Cain Dept. of Chemical Engineering, Louisiana State Univ., Baton Rouge, LA 70803.



**Fig. 1.** Idealized dredge-particle generating device and possible pathways for fate of contaminated sediment

Due to these losses to environmental media, dredging cannot remove all the contaminated material from a site.

Because the magnitude of the various losses is not well known, nor have previous attempts been made to quantify them, the objective of this paper is to present a multimedia model for mud and the associated chemical losses generated during dredging and to apply the model to the Bayou Bonfouca Superfund site, where polyaromatic hydrocarbon (PAH)-contaminated creosote waste was dredged.

## **General Model Approach**

The multimedia model developed is a steady-state box model consisting of many process elements, including the dredge element, which initiates chemical release in a flowing stream. The dredge delivers particles, and chemical release occurs after particles are resuspended in the water column. Chemicals bound to particles and in solution follow different pathways, as shown in Fig. 1; those bound to particles are advected downstream and redeposited, while those in solution are advected downstream as well, or evaporate to the air. Nevertheless, the majority of the chemical extracted is delivered to shore, although the exact quantity depends on many operational and site-specific factors.

The processes of advection and evaporation are losses since they depart the dredge area. The advection loss depends upon the type of silt containment employed. Of the two types allowed, the first is a curtain that is 100% efficient, in which all solids are retained while water is allowed to pass through; the solids retained are returned to the stream bottom as "solids fallback." The second scenario is a 0% efficient curtain, where all the solids generated move downstream and away from the dredge site.

Prior to dredging, an original amount of contaminated chemical exists in place. The mass of contaminated chemical is the product of mass of material and concentration averages of core samples obtained from the bed. Numerous core samples representing the vertical and areal extent of contamination allow the in-place contaminant mass to be estimated. This total amount, however, is not always targeted for removal since it may be impossible to reach and/or extract all contaminated material for a host of practical reasons. Therefore, the nontargeted mass of mud and chemical must be accounted for in the overall mass balance. For instance, in the Bayou Bonfouca case, any mud of total PAH lower than 1,300 mg/kg was nontargeted.

After dredging, the amount of contaminant redeposited in the sediment bed (fallback) plus the nontargeted mass constitutes the

unrecovered or remaining mass. The overall efficiencies of the dredging operation, defined as the amount of mud or contaminant delivered to shore divided by the original mass in place, are  $E_{\rm mud}$ % and  $E_{\rm chemical}$ %, respectively. Before the amount of each delivered to shore can be determined, it must be quantified. This is accomplished by performing an overall mass balance around the dredge area in the following manner: original contaminant in-place must equal the losses, the fallback, the nontargeted mass, and that delivered to shore. In order to compute these quantities, a solids and a chemical mass balance must be applied around the dredging operable unit. Typically this is a specific bottom area of the waterbody targeted for remediation by dredging.

## Solid-Particle Mass Balance

Solid particles are redistributed in the water column due to sediment resuspension by the dredge. The amount resuspended depends upon the type of dredge used. Detailed discussion of the types of dredges and dredging operations appear elsewhere (USEPA 1996).

The mass rate of sediment resuspended is proportional to the turbidity generated by the dredge. The so-called turbidity generating units (TGU) are used to define the mass suspended per unit volume of material extracted. The concept of the TGU was developed by Nakai (1978), according to whom, "the diffusion of suspended material is influenced by tidal current, grain sizes and other soil conditions, as well as the irregularities in the dredge action and bottom configuration." The general form of the TGU equation is

$$m = (q \cdot \text{TGU})/(R_0/R_{74})$$
 (1)

where *m* and *q*=mass and volume rate of sediment resuspension;  $R_0$ =fraction of sediment with a critical resuspension velocity less than the ambient current velocity; and  $R_{74}$ =fraction of sediment with diameter less than 74 microns.  $R_0/R_{74}$  is a fraction introduced to standardize the TGU because soil particles with diameters larger than 74 microns are not resuspended (Nakai 1978). Because the TGU depends upon the dredge operation and the bottom sediment characteristics, a wide range of TGU values exist. Nakai (1978) estimated the TGU by calculating the quantity of turbidity passing through a line of water sampling points. His results appear in Table 1 and are used in the multimedia model.

A dredging operable unit (DOU) is defined as a specific bottom area,  $A_D$  (m<sup>2</sup>), targeted for extraction operations. The limits of the area may or may not be physically defined by a silt curtain. With an average water depth, h(m), a water compartment or box is defined. The extraction operations occur within this volume, and there is continuous water movement through it,  $O(m^3/s)$ . The specific character of the location being dredged will determine how the DOU is defined. For example, it may be the exact boundaries of a well-characterized hot spot area near an outfall. Alternatively, it may be defined by economics, in which case limited funds are available for dredging, and the area targeted for extraction is projected accordingly. The DOU size may be defined by the season of the year in which such activities can occur. If the dredging project is particularly large in time and space, activities may be scaled to DOU sizes chosen so as to minimize ecosystem impact. For example, dredging may be discontinued during the fish-spawning season.

In order to perform a solids mass balance around the dredging operation "box", the dredge is assumed to operate within a volume of water,  $V_w$  (m<sup>3</sup>). A solids mass balance (in kg/s) performed around the water column portion of the box but excluding the sediment layer gives

Table 1. Turbidity Generating Units (TGUs) for Different Dredges and Dredging Projects (Nakai 1978)

Type of dredge	Installed power . or bucket volume		TGU		
		$(d < 74\mu, \%)^{a}$	$(d < 5\mu, \%)$	Classification <sup>b</sup>	(kg/m <sup>3</sup> )
Pump	4,000 hp	99.0	40.0	Silty clay	5.3
		98.5	36.0	Silty clay	22.5
		99.0	47.5	Clay	36.4
		31.8	11.4	Sandy loam	1.4
		69.2	35.4	Clay	45.2
		74.5	50.5	Sandy loam	12.1
	2,500 hp	94.4	34.5	Silty clay	9.9
		3.0	3.0	Sand	0.2
	2,500 hp	2.5	1.5	Sand	3.0
		8.0	2.0	Sand	0.1
Trailing suction	2,400 hp	92.0	20.7	Silty clay loam	7.1
	$\times 2$	88.1	19.4	Silty loam	12.1
	1,800 hp	83.2	33.4	Silt	25.2
Grab	8 m <sup>3</sup>	58.0	34.6	Silty clay	89.0
	$4 \text{ m}^3$	54.8	41.2	Clay	84.2
		45.0	3.5	Silty loam	15.8
	$3 \text{ m}^3$	62.0	5.5	Silty loam	11.9
		87.5	6.0	Silty loam	17.1
Bucket		10.2	1.5	Sand	17.6
		27.2	12.5	Sandy loam	55.8

 $^{a}d$  = diameter of soil particles.

<sup>b</sup>Classification is according to triangular soil classification system.

$$C_{SS}^{0} \cdot Q + \mathrm{TGU} \cdot V_{R} = C_{SS} \cdot Q \tag{2}$$

where  $C_{SS}^0$ =inlet solids concentration in solution (kg/m<sup>3</sup>);  $V_R$ =volumetric rate of sediment extracted (m<sup>3</sup>/s), and  $C_{SS}$ =suspended solids concentration in the box. Eq. (2) is used to estimate the suspended solids concentration, whether a silt curtain is employed or not.

The model allows for two particle-containment barrier scenarios. These semipermeable barriers, usually fabric-type devices called silt curtains, are placed both upstream and downstream of the dredge as an aid in preventing suspended solids from leaving the area. The first scenario assumes a 100% efficient silt curtain where the mass of fallback is derived from the TGU and the volume excavated. Upon settling within the box, it forms a layer of thickness,  $h_F$  (m). A simple mass balance yields the following for the thickness of the fallback layer:

$$h_F = (\mathrm{TGU} \cdot V) / (A_D \cdot \rho_b \cdot 2) \tag{3}$$

where  $A_D$ =surface area of the box; and V=dredging volume. A factor of 2 is added to the denominator of Eq. (3) because it is assumed that half of the fallback falls in front of the dredge as it moves across the bottom surface of the box.

The second scenario considered is that the silt curtain is 0% efficient. Here it is assumed that no solids are retained within  $A_D$  so that no fallback occurs. The dredge-generated suspended solids move downstream to points far afield and outside the box to become a lost quantity. The discussion of silt containment scenarios completes the solid particle-fate mass-balance portion of the multimedia model. Eqs. (1) through (3) capture the processes of particle generation, fallback, deposit formed by fallback, and advection away by flow.

#### **Chemical Mass Balance**

A chemical balance must be performed around the box containing the dredge element. Initially, the sediment bed is assumed to have an average chemical mass concentration  $\omega_c^0$  (mg/kg). If the mass of contaminated sediment is known, M (kg), the initial mass of chemical is

$$M = M_s \cdot \omega_c^0 \tag{4}$$

The quantities  $M_s$  and  $\omega_c^0$  are usually estimated based on core samples obtained throughout the box area  $A_D$  to define the depth, area, and level of contamination.

## **Chemical Partitioning within Suspended Mud Cloud**

During the operation of the dredge, particles become suspended in the water column. In order to relate the concentration of the particle-bound chemical ( $\omega_c$ ) and the chemical in solution ( $C_w$ ), a partition coefficient approach was used. Numerous investigators have studied the validity of the local equilibrium assumption (LEA) during sediment resuspension events and most conclude that it is unrealistic. Atkinson et al. (1995) compared suspension times for sediments of various sizes with expected equilibration times for desorption. They found that the equilibrium assumption was not valid for hydrophobic organic chemicals with partition coefficients higher than  $10^4$  mL/g because their equilibration time (1 to 10 days) exceeded their expected resuspension time. Brunck et al. (1993) measured mass transfer rates of tetrachlorinated dibenzo-dioxin/furans (TCDD/F) from suspended sediment particles to test the validity of a proposed equilibrium model. Their results indicated that equilibrium partitioning was inadequate for predicting TCDD/F fate in the environment because of mass transfer resistance and dissolved organic carbon releases from the solids. DePinto et al. (1998) performed model simulations on the desorption rates of two PCB congeners from suspended sediments, which suggested that a two-compartment equilibrium diffusion model fitted the data better than a single retarded nonequilibrium pore diffusion model. DiGiano et al. (1993) employed an

equilibrium-partitioning model that accurately predicted the concentration of soluble and sorbed PCB in the dredging elutriate test.

Due to the potential weakness of the LEA, it is more appropriate to employ an effective partition coefficient  $K_{de}$  for each molecular species modeled. Given the preliminary nature of the multimedia model, it is appropriate to employ such an approach. The use of  $K_d$  can represent the fact that equilibrium is unlikely to be reached. For example, if it has been measured, only the readily desorbable or labile fraction  $(f_e)$  that responds to equilibrium is used. In addition, the fraction value of the desorption partition coefficient  $(f_d)$  may be less than the typical adsorption coefficient reported in the literature. In both cases these fractions are multiples of the LEA coefficient so that it can be transformed by the fate product to yield  $K_{de}$ .

A mass balance on chemical A around the box containing the dredge element can now be performed (mg/s):

$$(C_{SS}^{0} \cdot \omega_{A}^{00} \cdot Q) + (C_{w}^{0} \cdot Q) + (\mathrm{TGU} \cdot V_{R} \cdot \omega_{A}^{0})$$
  
=  $(\mathrm{TGU} \cdot V_{R} \cdot \omega_{A} \cdot f) + [(C_{SS} \cdot \omega_{A} \cdot Q) \cdot (1-f)] + (C_{w} \cdot Q)$   
+  $[K_{e} \cdot (C_{w} - C_{a}/H) \cdot A_{D}] + (A_{D} \cdot h \cdot C_{w} \cdot K_{rx})$  (5)

The three inflow terms on the left side are advective inflow of A on suspended particles and in solution, followed by the mass of A delivered into the water column by action of the dredge resuspending particles. The outflow terms are fallback of particles containing A, particles containing A departing with the water currents, A departing in solution with water currents, evaporation through the air-water interface, and reaction loss of A in the water column. Using  $K_{de}$ , the concentration on suspended particles  $(\omega_c)$  and the chemical in solution  $(C_w)$  are related by means of a partition coefficient defined as follows:  $K_{de} = \omega_c / C_w$ . The evaporation term consists of an overall mass transfer coefficient,  $K_e$ ; concentration of A in solution,  $C_w$  (mg/m<sup>3</sup>); concentration of A in the air above the box,  $C_a$  (mg/m<sup>3</sup>); and the Henry's constant for chemical A, H (m<sup>3</sup> H<sub>2</sub>O/m<sup>3</sup> air). The parameter f is a factor that accounts for the efficiency of the silt curtain. If the curtain is 100% efficient, f=1 and fallback occurs (no particle advection downstream); whereas if the curtain is 0% efficient, f=0 and advection of chemical in particles downstream occurs (no fallback). The first-order reactive decay term for chemical in the water is  $K_{rx}$  in s<sup>-1</sup>;  $K_{rx} = 0.693/\tau_{1/2}$ , where  $\tau_{1/2}$  is the decay half-life.

Eqs. (1) through (5) are the working forms of the steady-state multimedia model. Typically, the desired computed unknowns are suspended solids concentration  $(C_{SS})$ ; fallback layer thickness  $(h_F)$ ; concentration of chemical in solution  $(C_w)$ ; and concentration on suspended particles  $(\omega_A)$ . Once these parameters are known, they contain the information necessary to calculate contaminant mass losses. In effect, the multimedia compartmental box model is a near-field fate and transport model whose purpose is a first-order estimate of chemical fate near the dredge device. Other models exist that are helpful in projecting particle and possibly chemical dispersion and fate farther from the dredge operation (Hayes and Kuo 1991). In addition, the Corps of Engineers has developed a steady-state "DREDGE" software to estimate the resuspension and transport of sediments and associated chemicals during dredging operations. The software combines empirical sediment resuspension (near-field) models and simple suspended sediment transport (far-field) models to estimate suspended sediment concentrations at specified water column locations (Hayes et al. 1996). In a fashion similar to the multimedia model, "DREDGE" uses an equilibrium approach to predict dissolved contaminant concentrations from suspended solids concentrations. However, there are many differences between the existing models and the near-field model proposed here, mainly because other release processes such as evaporation, advection, and fallback can be quantified, which in turn allows effectiveness to be defined and evaluated.

## Model Inputs and Rate Processes

Numerous parameters are required as inputs to the model. These are listed in the Appendix. Some are obtained from field measurements, and others from published sources of data and information. Compound-specific properties such as Henry's constant, partition coefficients, and molecular diffusivities are readily available from numerous literature sources. The evaporation mass-transfer coefficient  $K_e$  can be estimated using the two-resistance theory of interphase mass transfer, which relates the overall mass transfer coefficient,  $K_e$ , to the individual liquid- and gas-phase coefficient as follows:

$$(1/K_e) = (1/k_l) + (1/(k_g \cdot H)) \tag{6}$$

where  $k_l$  (m/s) is the local liquid-phase mass-transfer coefficient and  $k_g$  (m/s) is the local gas-phase mass-transfer coefficient. The liquid-phase coefficient is the sum of the contributions of processes that induce turbulence in the surface waters. These include the hydraulics of water flow, the wind-induced component, and that induced by the mechanical dredge. Dredges that operate without continuously disturbing the air/water interface, such as the cutterhead and auger dredges, are not significant factors in the evaporation process and are not included in this model development.

Accepted correlations and procedures are used for estimating the hydraulic water flow and wind-induced evaporation transport coefficients (Thibodeaux 1996). Without specific data, the mechanical dredge contribution must otherwise be estimated. For modeling purposes, it will be assumed that the dredge bucket acts much as a mechanical surface agitator. These are designed to stir the liquid surface, and the "bucket" of a mechanical dredge behaves similarly as it moves up and down through the surface. A correlation for the liquid-phase mass transfer coefficient for oxygen absorption is used as follows [the original correlation for oxygen absorption modified for volatile species is (TPC 1971)]:

$$k_l = 3,140 \cdot (D_A / D_B)^{1/2} \cdot (n \cdot E \cdot \alpha) \cdot (1.024)^{(T - 20^{\circ}\text{C})}$$
(7)

where the coefficient is in centimeters per hour (cm/h); D, the molecular diffusivity of chemical (A) and oxygen (B) respectively; and T=water temperature in degrees Celsius. The plane surface area,  $A_d$ , for an aerator-equivalent dredge that delivers 10 kW of power (13.4 hp), P, or greater is (Thibodeaux 1996):

$$A_d = 17 \cdot P \tag{8}$$

where the area is in square feet. The following equivalent parameters are needed in Eqs. (7) and (8): n=2 to 4 lb O<sub>2</sub>/h-hp. Oxygen delivery of 2 is suggested because bucket dredges are likely not the most efficient aerators; however, they create turbulence through the water column, whereas surface aerators stir the surface only. The *E* (aerator power delivery efficiency) ranges from 0.65 to 0.9. The lowest value of 0.65 was used because bucket dredges do not spend the entire time in the water; thus efficiency is likely to be low. The value for *P* [the nameplate horsepower (hp)] recommended by the manufacturer of the dredge should be used. The  $\alpha$  (dirty-water to clean-water ratio) ranges from 0.8 to 0.85; a conservative value of 0.8 was used because the water surface usually appears oily once dredging is under way.

The resulting coefficient-area product needed for Eqs. (7) and (8) must be corrected because the volatilization surface area is not the surface area of the entire site, but only the surface area  $A_d$  agitated by the dredge. The following correction is used:

$$k_{l(\text{corrected})} = k_l \cdot (A_d / A_D) \tag{9}$$

where  $A_D$ =cordoned-off or otherwise designated area of the dredge site. Once obtained, the water, wind, and dredge-induced evaporation mass-transfer coefficients are summed in order to estimate their combined effects.

For closure on Eq. (6), a gas-phase coefficient is needed; a correlation for water evaporation from lakes is used (Thibodeaux 1996):

$$k_g = 358 \cdot v_w \cdot A_D^{-0.05} \tag{10}$$

where  $k_g$  is in centimeters per hour (cm/h);  $v_w$ =wind speed (in miles per hour); and  $A_D$ =surface area of the site in acres.

## **Contaminant Losses and Dredging Effectiveness**

Dredging produces contaminant losses to the surroundings due to the unconfined nature of the operation. Once the suspended sediment and chemical concentrations are obtained, the process losses due to advection (both particle and soluble), evaporation, and solids fallback can be estimated. Once these individual losses are determined, an overall mud-layer mass balance is performed as follows to determine the amount of mud and contaminant delivered to shore: original quantity of contaminant in place must equal the losses, the fallback, and the nontargeted mass and that delivered to shore. The original volume or mass of contaminated mud and chemical species in place is typically estimated from field data that represent area and depth averaged concentrations from bottom core samples. The model user must specify the target mud volume as an area  $A_D$  and a depth h. Depending on the efforts devoted to characterizing the site, the magnitude of this volume may be known with various degrees of certainty. In the case of a planned dredging operation, certain other parameters need to be estimated as well.

The evidence gathered to date on dredging operations suggests that significant quantities of the targeted mass cannot be recovered (Hudson Voice 2001). The reasons are numerous and include in-water and on-bottom obstructions that preclude effective mud extraction and deficiencies in design or operations of the equipment. These published case studies can be used to estimate the design-unextractable quantities. Frequently pre- and postdredging bathymetry is used to quantify this mass. Resampling and analysis of bottom cores are required for assessing the remaining chemical mass. In some design cases, such as Bayou Bonfouca, a level of concentration is set and the mass on bottom at or below this level is not targeted for extraction. The mud and the chemical balance differ in the loss quantities. There are no evaporation or advective solute components in the mud balance. With these various masses quantified, the overall efficiencies of the dredging operation,  $E_{\rm mud}\%$  or  $E_{\rm chemical}\%$  , defined as that delivered to shore divided by original mass in place, can be obtained as well as other effectiveness measures.

## **Application to Bayou Bonfouca**

The multimedia model was applied to quantify the losses and determine the overall efficiency of the dredging operation for the Bayou Bonfouca Superfund site in Slidell, Louisiana. The site is located adjacent to the abandoned American Creosote Works plant 7 mi north of Lake Pontchartrain, with the bayou running along the south side of the plant (USDHHS 1992). Bayou Bonfouca is a tidal-influenced fresh water estuary with an estimated net discharge rate of 13 ft<sup>3</sup>/s. It is about 10 ft deep, with an average width of 125 ft. The contaminants of concern are polynuclear aromatic hydrocarbons (PAHs), which account for 90% of the 400 components found in creosote (CH2M Hill 1986). Among the PAHs are six known carcinogens: benzo(a)pyrene, benzo(a)anthracene, benzo(b)fluoranthene, benzo(k)fluoranthene, indeno(1,2,3-cd)pyrene, and chrysene. Aside from the six carcinogens, there are 12 other contaminants of concern that make up the total PAH mixture.

The American Creosote Works began production of creosote and the treatment of various woods in 1892. Production ceased in 1972 when a fire destroyed the plant. The creosote contamination of the bayou was the result of industrial process operations, disposal practices, and occasional spills that occurred over time. Dredging in the bayou commenced in March 1994 and was completed in about a year. A bucket dredge consisting of a P984 Liebherr backhoe with a 5 cu yd excavation bucket was used to remove the contaminated sediment in the bayou (Bean 1993). The dredging operation in Bayou Bonfouca, conducted by the Corps of Engineers, is one of the largest and costliest environmental dredging projects to date (\$130 million) (USEPA 1997).

Information obtained about this dredging operation is needed to illustrate the application of the multimedia model and the types of information it is capable of providing. Much data at a particular site need to be collected during and after dredging in order to compare the predictions of this multimedia model to a real-world operation. In the case of the Bayou Bonfouca dredging operation and most others, such data collection is absent. Although the model results are hypothetical, they do provide mass-balance information, relative magnitudes of the various losses, and some sense as to the expected efficiency of the overall extraction operation. So, what follows are hypothetical mud and contaminant fate projections for a real-world dredging operation that was performed in the mid-1990s.

## **Results and Discussion**

Values for the input parameters for Bayou Bonfouca are presented in Table 2. The model quantifies the losses that occur during dredging. Even though some additional losses such as evaporation and advection will occur during the nonoperation time, they are neglected. Typically the operation period is 8 h per day, 5 days a week. If a 24 h period is used, this will lead to overestimation of evaporation because, without a generation source, the concentration and rate decreases until the dredging restarts. In addition, the model would not be consistent with other losses such as fallback, which only occurs while the dredge is operating.

The mud balance results appear in Figs. 2(a) and 2(b). A total of 2.18E8 kg of contaminated sediment was targeted in the bayou. This value is three orders of magnitude greater than the mass of total chemical targeted (7.4E5 kg). Of the 2.18E8 kg of contaminated sediment targeted, 3.65E7 kg were nontargeted, which corresponds to 14.3% of the total amount of mud in place. Furthermore, the mud losses are not very significant. When the mud is primarily sand (TGU=17.6 kg/m<sup>3</sup>), only 0.91 and 0.45% of the total mud targeted was lost to advection and fallback, respectively, for silt curtain 0 and 100% efficient, respectively. Con-

Table 2.	Multimedia	Model	Inputs	for	Bayou	Bonfouca

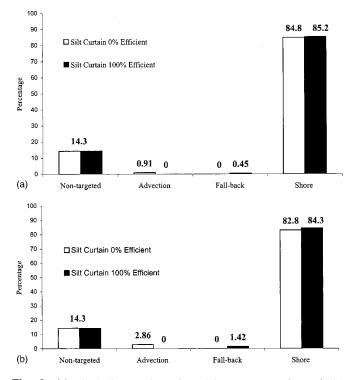
Parameter	Amount	Definition	
$C_{SS}^0$	6.82 kg/m <sup>3</sup>	Based on reported bayou data	
		(IT-OHM 1993)	
TGU	17.6 kg/m <sup>3</sup>	Observed TGU values for a bucket dredge	
	55.8 kg/m <sup>3</sup>	operating in sand and sandy loam	
		(Nakai 1978)	
Q	$0.365 \text{ m}^3/\text{s}$	Bayou flow-through data (USGS 2000)	
V	130,000 m <sup>3</sup>	Reported data (USEPA 1997)	
$t_D$	7,488,000 s	Based on 40 h weekly dredging period for	
		52 weeks	
$A_D$	40,650 m <sup>2</sup>	Calculated from blueprints	
		(CH2M Hill 1990).	
$\rho_b$	$1,680 \text{ kg/m}^3$	Same as deep sediment density. Based on	
		water content of 52% and sediment dry	
		density of 2.6 g/cm <sup>3</sup> (CH2M Hill 1988)	
$\omega_c^0$	3,380 mg/kg	Estimated from data for each PAH (USEPA	
		1989)	
$\omega_c^{00}$	0 mg/kg	Inflow concentration of chemical in particles	
		assumed to be negligible	
$C_w^0$	0 mg/m <sup>3</sup>	Assumed to be zero because chemical was	
		not brought to bayou from a source upstream	
$C_a$	0 mg/m <sup>3</sup>	No measurements of chemical concentrations	
		in background air were available. Assumed	
		to be zero to provide for worst case scenario	
<i>v</i> <sub>w</sub>	4.9 m/s	Based on USGS (2000) data	

versely, when the mud is sandy loam (TGU=55.8 kg/m<sup>3</sup>), the losses are greater: 2.86 and 1.42%.

The TGU values reported by Nakai (1978) for a bucket dredge operating in sand and sandy loam were employed for comparison. As the TGU increases, the multiphase output concentrations as well as the suspended solids concentration ( $C_{SS}$ ) around the dredge zone also increase. Furthermore, the fallback layer thickness ( $h_F$ ) also increases. This is reasonable because the TGU is a measure of the turbidity generated by the dredge. The higher the TGU, the higher the amount of chemical resuspended. Chemical resuspension in turn leads to dispersion of contaminants to all phases (air, water, sediment, and particles).

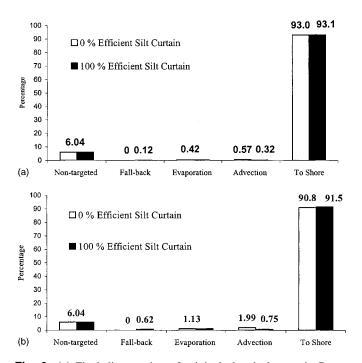
The chemical results appear in Figs. 3(a and b). Contaminant losses are directly proportional to the computed solid (TSS) and chemical concentrations. The fate of a contaminant is also affected by its chemical properties. This behavior is shown in Figs. 4(a) and 4(b), in which the percent loss of each of the processes quantified by the model is plotted versus its correspondent PAH in order of increasing octanol-water partition coefficient  $(K_{ow})$ . The  $K_{ow}$ , defined as the ratio of the molar or mass concentrations in the water-saturated octanol and the octanol-saturated water phase, is a measure of a compound's hydrophobicity. That is, the higher the  $K_{ow}$ , the higher the affinity for a compound to associate with the solid phase. This trend can be observed with the 16 PAHs present in Bayou Bonfouca. As the  $K_{ow}$  increases, the percent losses due to fallback and particle advection rise, while those percent losses associated with evaporation and soluble advection decrease progressively to almost zero. The  $K_{ow}$  range among the PAHs studied varies from 3.30 ( $\log K_{ow}$ ) for naphthalene (Campbell and Luthy 1985) to 6.70 for indeno(1,2,3-cd)pyrene (Doucette et al. 1988). Next the results of 2 of the 16 PAHs modeled are compared.

Naphthalene, the PAH with the highest concentration in Bonfouca (32.5%), is very soluble ( $C_w = 263.2 \text{ mg/m}^3$ ) and has the

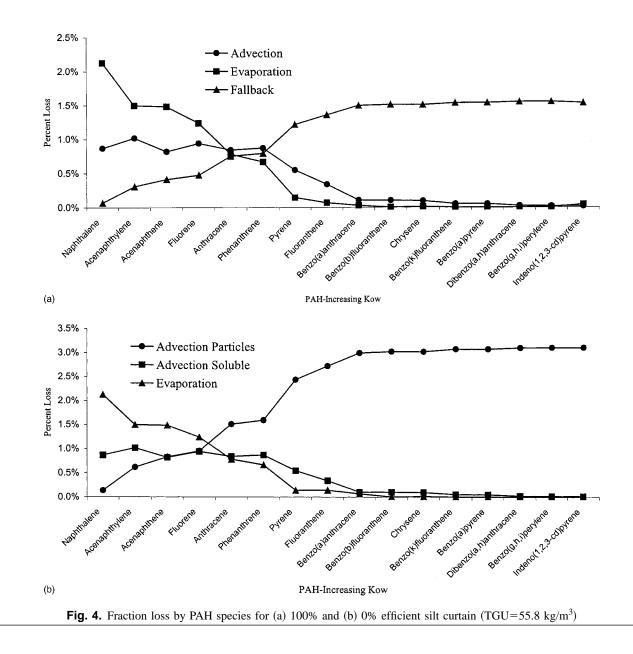


**Fig. 2.** (a) Final dispensation of mud in Bayou Bonfouca (TGU =  $17.6 \text{ kg/m}^3$ ) (b) Final dispensation of mud in Bayou Bonfouca (TGU= $55.8 \text{ kg/m}^3$ )

highest evaporation mass-transfer coefficient of all the PAHs  $(K_e = 2.2E - 05 \text{ m/s})$ . Because of this, naphthalene evaporates very readily and also leaves in solution. Conversely, naphthalene has the lowest partition coefficient of all the PAHs  $(K_D = 0.06 \text{ m}^3/\text{kg})$  and therefore is not very persistent in the solid



**Fig. 3.** (a) Final dispensation of original chemical mass in Bayou Bonfouca (TGU=17.6 kg/m<sup>3</sup>) (b) Final dispensation of original chemical mass in Bayou Bonfouca (TGU=55.8 kg/m<sup>3</sup>)



phase. Comparing the naphthalene losses for a 100% efficient silt curtain when TGU=17.6 kg/m<sup>3</sup>, the results show that 18 kg of naphthalene were lost to fallback. This is negligible compared to the 1,764 and 720 kg of naphthalene that were lost to evaporation and soluble advection. For the case of 0% efficient silt curtain (no fallback), 36 kg of naphthalene advected out in particles. This is also small compared to the amount evaporated and advected out in solution (1,764 and 720 kg, respectively). This result is consistent with the high solubility of naphthalene.

Next, benzo(a)pyrene will be contrasted to naphthalene. This PAH, which only accounts for 0.7% of the total PAH mixture, is sparingly soluble ( $C_w = 0.95 \text{ mg/m}^3$ ). In addition, it has an evaporation mass transfer coefficient 3 orders of magnitude smaller than naphthalene ( $K_e = 9.35\text{E}-08 \text{ m/s}$ ). Because of its low solubility, benzo(a)pyrene is highly persistent in the solid phase and mostly falls back onto the bottom for a 100% efficient silt curtain, or leaves in particles for a 0% efficient silt curtain. Only 0.03 kg is lost to evaporation. This is very significant because benzo(a)pyrene is a known carcinogen, along with benzo(a)anthracene, benzo(b)fluoranthene, benzo(k)fluoranthene, indeno(1,2,3-cd)pyrene, and chrysene (USDHHS 1992). It is desirable that these carcino-

gens remain in the sediment or in particles rather than in solution or in the air because exposure to humans is more likely to occur by means of water contact or intake and air inhalation (breathing). The general trend of the carcinogens present in Bonfouca is the same; that is, they are not very soluble, nor do they evaporate very readily. Instead, they have high partition coefficients and therefore tend to remain in the solid phase.

In order to compare the overall multimedia losses, it is important to first know or quantify the amount or total amount of chemical present prior to dredging. This amount is termed "total in-place", and it is made up of the targeted plus the nontargeted amount. In Bayou Bonfouca, 786,000 kg of total PAH was inplace, of which 738,000 kg was targeted and 47,500 kg was nontargeted. The nontargeted amount corresponds to 6.04% of the total amount in place. Of the targeted mass, 3,300 kg was lost to evaporation and 4,440 kg was lost to advection for a 0% efficient silt curtain. These masses correspond to only 0.42 and 0.57% of the total in place, respectively. For the case of a 100% efficient silt curtain, 3,310 kg was lost to evaporation, while the 2,540 kg lost to advection was roughly half compared to the 0% efficient silt curtain (recall no chemical advects out in particles). However, some chemical was lost to fallback (946 kg; only 0.12% of the total in place). These results for a TGU=17.6 kg/m<sup>3</sup> (sandlike sediment) are summarized in Fig. 3(a).

For sandy loam (TGU=55.8 kg/m<sup>3</sup>), the multimedia losses are higher. For instance, 8,900 kg of total PNAs evaporated, while 15,700 kg advected out in both particles and solution. This corresponds to 1.13 and 1.99% of the total in place, respectively. Because the multimedia losses are higher for sandy-loam type sediment, the amount of total contaminant delivered to shore will be less. Of the total amount in place, 90.8 and 91.5% of the chemical goes to shore when the curtain is 0 and 100% efficient, respectively. In contrast, 93.0 and 93.1% of the total chemical in place is delivered to shore when the sediment is sand (TGU=17.6  $kg/m^3$ ). This result suggests that the dredge is more efficient when it operates in sand rather than sandy loam. Fig. 3(b) summarizes the final dispensation of original contaminated material in Bayou Bonfouca when TGU=55.8 kg/m<sup>3</sup>.

During the remediation operation in Bayou Bonfouca, limited concentration measurements were available during and after dredging in the bed surface and water column. As a result, field data such as TSS and PAH water-column concentrations during dredging and capping were not available to test model predictions. However, two surficial grab samples were obtained on top of the cap in September 1997 (LDEQ 1997). Even though the two samples collected were expected to register nondetect suspended particle concentrations,  $(\omega_c)$ , because of the clean cap, detectable PAH concentrations were found. The two collected samples registered concentrations of 5 and 25 mg/kg total PAH. These values appear low, but this is due in part to the clean cap placed in the bayou after the dredging operation concluded in March 1995. In addition, the samples were collected 2 years after dredging took place, and the PAHs were likely "weathered" and diluted with the clean material. The model predicted higher suspended particle concentrations ranging from 823 to 1,340 mg/kg total PAH, depending on the TGU employed and the efficiency of the silt curtain. The predicted values reflect the total PAH concentration immediately after dredging is halted, and not 2 years later.

In order to assess dredging effectiveness, some estimates were made on concentration and mass reductions. For instance, 62% concentration reduction occurred from the predredge average of 3,380 mg/kg total PAH to the 1,300 mg/kg target. Based on bathometric measurements and bed-profile concentrations, it was estimated that 86% of the contaminated sediment mass was removed, along with 94% of the PAH mass (Duckworth 1999). This result is consistent with the model, which predicted that the dredge would remove and deliver 91 to 92% of the total contaminant mass to shore when the sediment is sandy loam (TGU=55.8  $kg/m^3$ ), and around 93% when the sediment is sand (TGU=17.6  $kg/m^3$ ).

#### Summary and Closure

The use of dredging in remediating contaminated sediment beds requires a means of forecasting the magnitude of chemical losses associated with the type of dredge selected and other particular characteristics of the site. A multimedia compartmental chemical fate and transport model was developed to simulate and quantify the most significant processes triggered during bed excavation. For a defined dredging operable unit (DOU), these included particle generation, release in soluble and particulate form, advection, evaporation, reaction, and fallback. Based on the initial inplace chemical mass, the fractions lost were combined with the

nontargeted and unextractable mass to quantify the mass delivered to shore. The latter is used as an overall measure of dredging effectiveness, defined as the percent ratio of the quantity delivered to shore to the original in-place quantity.

As an illustration, the model was applied to the Louisiana Bayou Bonfouca site, which was dredged in 1995. A mechanical dredge was employed to extract 130,000 m<sup>3</sup> of PAH contaminated sediment. Specific site conditions were used in order to project the magnitude of the losses encountered in a large environmental dredging project and to investigate the sensitivity of a few input parameters suspected to have a significant effect on the magnitude of the losses. Two were highlighted: the particle loss rate of the dredge, called TGU, and the use or absence of silt curtains to contain the mud cloud generated. The estimates produced by the model were that 90 to 93% of the in-place PAH mass was delivered to shore in all cases, and 6% was nontargeted for removal. Very small percentages of the in-place PAH mass were lost, with or without the use of silt curtains.

Advection was the largest loss process. The model output was 0.57 and 1.99% for the 17.6 and 55.8 TGU values, respectively. Evaporation at 0.42 to 1.13% was next, while fallback was 0.12 to 0.62%. The reader should appreciate that although the percent losses appear small in comparison to the initial in-place mass and mass delivered to shore, it is the mass quantity lost that impacts the surrounding ecosystem. Both downstream and off-site chemical releases via the water and air routes are additional contributions to any ongoing releases to these media. In the case of Bayou Bonfouca, the advection and evaporative losses contributed up to 15,666 and 8,913 kg total PAHs, respectively, for the 1 year dredging period.

Although the model is based on mass-balance principles and contains chemodynamic process algorithms used in similar contexts, it is a first attempt at developing one. In addition, the losses and extractions are reasonable and in line with the limited available data. Projections made by the model are entirely hypothetical since many of the model assumptions and process components used need further study, development, and evaluation. Its use at this time should be for screening purposes only since it lacks any real verification or comparison to field data.

#### Acknowledgment

This work was supported by funds provided by the Louisiana State University Gordon A. and Mary Cain endowment in the Department of Chemical Engineering.

## Notation

The following symbols are used in this paper:

- $A_D$  = dredging area (m<sup>2</sup>);
- $A_d$  = surface area agitated by dredge (m<sup>2</sup>);
- $C_a$  = concentration of species A in background air  $(mg/m^{2});$
- $C_{SS}$  = suspended solids concentration (kg/m<sup>3</sup>);  $C_{SS}^{0}$  = inflow solids concentration (kg/m<sup>3</sup>);
- $C_w$  = concentration of species A in solution (mg/m<sup>3</sup>);
- $C_w^0$  = inflow concentration of species A in solution (ling)in  $C_w^0$  = inflow concentration of species A in solution  $(mg/m^3);$
- $D_A$  = molecular diffusivity of species A in water  $(m^2/s);$
- $D_B$  = molecular diffusivity of species B in water  $(m^2/s);$

- E = aerator power delivery efficiency (dimensionless);
- $E_{\text{chemical}} = \text{overall chemical removal efficiency (dimension-less);}$ 
  - $E_{\text{mud}}$  = overall mud removal efficiency (dimensionless);
    - f = silt curtain efficiency factor (dimensionless);
    - H = Henry's constant of species A (m<sup>3</sup> H<sub>2</sub>O/m<sup>3</sup> air);
    - h = water depth (m);
    - $h_F$  = fallback layer thickness (m);
  - $K_{de}$  = effective partition coefficient between solids and water (m<sup>3</sup>/kg);
  - $K_e$  = evaporation mass transfer coefficient (m/s);
  - $k_g$  = local gas-phase mass transfer coefficient (m/s);
  - $k_l = \text{local liquid phase mass transfer coefficient (m/s);}$
  - M = initial mass of total chemical (kg);
  - $M_s$  = mass of contaminated sediment (kg);
  - m = mass rate of sediment resuspended (kg/s);
  - $n = \text{oxygen delivery (lb O}_2/\text{h-hp});$
  - P = dredge nameplate horsepower (hp);
  - Q = volumetric water flow rate (m<sup>3</sup>/s);
  - q = volume rate of sediment resuspended (m<sup>3</sup>/s);
  - $R_0$  = fraction of sediment with critical resuspension velocity less than ambient current velocity (dimensionless);
  - $R_{74}$  = fraction of sediment with diameter less than 74  $\mu$ m (dimensionless);
  - T = water temperature (°C);
  - TGU = turbidity generating unit (kg/m<sup>3</sup>);
    - $t_D$  = dredging time (s);
    - $V = \text{dredging volume } (\text{m}^3);$
    - $V_R$  = volumetric rate of sediment extracted (m<sup>3</sup>/s);
    - $V_w$  = water volume (m<sup>3</sup>);
    - $v_w$  = wind speed (m/s);
    - $\alpha$  = dirty water to clean water ratio (dimensionless);
    - $\rho_b$  = bed-sediment density (kg/m<sup>3</sup>);
    - $\omega_A$  = concentration of species A on suspended particles (mg/kg);
    - $\omega_A^0$  = average concentration of species A in sediment bed (mg/kg);
  - $\omega_A^{00} = \text{inflow concentration of species A in particles}$ (mg/kg);
  - $\omega_c^0$  = average concentration of total chemical in sediment bed (mg/kg); and
  - $\omega_c^{00}$  = inflow concentration of total chemical in particles (mg/kg).

## References

Atkinson, J. F., Chen-Yu, C., and Depinto, J. V. (1995). "Desorption during resuspension events: Kinetic v. equilibrium model." *Mar. Freshwater Res.*, 46(1), 251–6.

Bean, T. E. C. (1993). Bayou dredging work plan, Slidell, La., May 18.

- Brunck, R., Hinton, S. W., and Walbridge, L. (1993). "Mass transfer of TCDD/F from suspended sediment particles." *Water Sci. Technol.*, 28(8-9), 181–90.
- Campbell, J. R., and Luthy, R. G. (1985). "Prediction of aromatic solute partition coefficients using the UNIFAC group contribution model." *Environ. Sci. Technol.*, 19(10), 180–185.

- CH2M Hill. (1986). *Bayou Bonfouca site remedial investigation (RI) rep., Final*, Slidell, La, April 25.
- CH2M Hill. (1988). Summary of design investigations, Bayou Bonfouca remedial design, Appendix A, Montgomery, Ala., December 9.
- CH2M Hill. (1990). Bayou Bonfouca source control remedial action, Blueprints, Slidell, La., August.
- Depinto, J. V., and Gong, Y. (1998). "Desorption rates of two PCB congeners from suspended sediments—II. Model simulation." *Water Resour.*, 32(8), 2518–2532.
- DiGiano, F. A., Miller, C. T., and Yoon, J. (1993). "Predicting release of PCBs at point of dredging." J. Environ. Eng., 119(1), 72–89.

Doucette, R. W., Dupont, R. R., Grenney, W. J., McLean, J. E., and Sims, R. C. (1988). "Treatment potential for 56 EPA listed hazardous chemicals in soil." U.S. EPA Rep. 600/6-88-001.

- Duckworth, K. (1999). "The effectiveness of environmental dredging: A study of three sites." MS thesis, Louisiana State Univ., Baton Rouge, La.
- General Electric Company (GEC). (2000). Environmental dredging: An evaluation of its effectiveness in controlling risks, Corporate Environmental Programs, Albany, N.Y., August.
- Hayes, D. F. (1992). "Anticipating sediment resuspension and contaminant release during environmental dredging operations." Proc., Int. Symposium on Environmental Dredging.
- Hayes, D. F., Je, C., and Tang, T. (1996). "Dredge module technical documentation." Dept. of Civil and Environmental Engineering, Univ. of Utah, Module of the Automated Dredging and Disposal Alternatives Modeling System (ADDAMS) distributed by the U.S. Army Corps of Engineers through the Environmental Laboratory, USAE Research and Development Center Waterways Experiment Station, Vicksburg, Miss.
- Kuo, A. Y., and Hayes, D. F. (1991). "Model for turbidity plume induced by bucket dredge." J. Waterwa., Port, Coastal, Ocean Eng., 117(6), 610–623.
- "Hudson Voice: Dredging PCBs in the Hudson River." (2001). (http:// www.hudsonwatch.com).
- IT-OHM. (1993). Bayou Bonfouca Bayou water quality monitoring background study report, Slidell, La., July.
- Louisiana Department of Environmental Quality (LDEQ). (1997). Bayou Bonfouca sediment and water samples 9/16/97, Semivolitile Organics by EPA method 8270, Technical Services Division Water Laboratory, Slidell, La.
- Nakai, O. (1978). "Turbidity generated by dredging projects." Proc., 3rd U.S./Japan Experts Meeting, U.S. Army Engineer Water Resources Support Center, Ft. Belvoir, Va.
- National Research Council (NRC). (1998). National symposium on contaminated sediments: Coupling risk reduction with sustainable management and reuse, Conf. Proc., Washington, D.C.
- Technical Practice Committee (TPC). (1971). *Manual of practice 5*, Subcommittee on Aeration in Wastewater Treatment, Water Pollution Control Federation, Washington, D.C.
- Thibodeaux, L. J. (1996). *Environmental chemodynamics*, Wiley, New York.
- U.S. Dept. of Health and Human Services (USDHHS). (1992). "Public health assessment for Bayou Bonfouca." *LAD980745632*, Agency for Toxic Substances and Disease Registry, Slidell, La.
- U.S. Environmental Protection Agency (USEPA). (1989). Design investigation rep.: Bayou Bonfouca source control operable unit, Region IV, prepared by CH2M Hill, for EPA, work assignment No. 160-6N38-0, CH2M Hill Master Project No. MGM69140.
- U.S. Environmental Protection Agency (USEPA). (1997). Final close out rep.: Source control operable unit, Bayou Bonfouca superfund site, Region VI, Superfund Division, Slidell, La.
- U.S. Geological Survey (USGS). (2000). Environmental data. (http://www.usgs.gov).