

# Planning-Level Source Decay Models to Evaluate Impact of Source Depletion on Remediation Time Frame

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A recent United States Environmental Protection Agency (US EPA) Expert Panel on Dense Nonaqueous Phase Liquid (DNAPL) Source Remediation concluded that the decision-making process for implementing source depletion is hampered by quantitative uncertainties and that few useful predictive tools are currently available for evaluating the benefits. This article provides a new planning-level approach to aid the process. Four simple mass balance models were used to provide estimates of the reduction in the remediation time frame (RTF) for a given amount of source depletion: step function, linear decay, first-order decay, and compound. As a shared framework for assessment, all models use the time required to remediate groundwater concentrations below a particular threshold (e.g., goal concentration or mass discharge rate) as a metric. This value is of interest in terms of providing (1) absolute RTF estimates in years as a function of current mass discharge rate, current source mass, the remediation goal, and the source-reduction factor, and (2) relative RTF estimates as a fraction of the remediation time frame for monitored natural attenuation (MNA). Because the latter is a function of the remediation goal and the remaining fraction (RF) of mass following remediation, the relative RTF can be a valuable aid in the decision to proceed with source depletion or to use a long-term containment or MNA approach. Design curves and examples illustrate the nonlinear relationship between the fraction of mass remaining following source depletion and the reduction in the RTF in the three decay-based models. For an example case where 70 percent of the mass was removed by source depletion and the remediation goal ( $C_g/C_0$ ) was input as 0.01, the improvement in the RTF (relative to MNA) ranged from a 70 percent reduction (step function model) to a 21 percent reduction (compound model). Because empirical and process knowledge support the appropriateness of decay-based models, the efficiency of source depletion in reducing the RTF is likely to be low at most sites (i.e., the percentage reduction in RTF will be much lower than the percentage of the mass that is removed by a source-depletion project). Overall, the anticipated use of this planning model is in guiding the decision-making process by quantifying the relative relationship between RTF and source depletion using commonly available site data. © 2005 Wiley Periodicals, Inc.

## INTRODUCTION

There is considerable debate about the effectiveness and appropriateness of source depletion (i.e., the removal of contaminant mass from the source zone) at sites with groundwater plumes. For example, the United States Environmental Protection Agency (US EPA) recently commissioned an Expert Panel on Dense Nonaqueous Phase Liquid (DNAPL) Source Remediation (Kavanaugh et al., 2003). The Panel was

charged with addressing several questions, including: (1) what are the potential benefits and the potential adverse impacts of DNAPL source depletion as a remediation strategy? (2) what performance can be anticipated from source-zone mass-depletion technologies? and (3) are currently available tools adequate to predict the performance of source-depletion options?

The Expert Panel concluded, “Quantitative predictions of these potential benefits and adverse impacts to aid decision making on whether to implement DNAPL source depletion actions are highly uncertain. These uncertainties remain as significant barriers to more widespread use of source depletion options” (Kavanaugh et al., 2003). The Expert Panel identified “the development of more user-friendly prediction tools that provide a more reliable and accurate basis for assessing the likely performance of source-zone depletion technologies” as a “primary research need.” While several researchers are working on improving our understanding of source zone response and source-zone modeling (e.g., Abriola et al., 2002; Barth et al., 2003; Gerhard & Kueper, 2003; Lemke et al., 2004; Rao et al., 2001; Wood et al., 2003), very few useful predictive tools are currently available for evaluating the benefits of source depletion.

This article provides a new planning-level approach for evaluating the benefits of partial source depletion (as complete source depletion is considered to be impossible at all but a few DNAPL sites). Four simple mass balance models are used to provide estimates of the reduction in the remediation time frame (RTF) for a given amount of source depletion.

## DEFINITIONS

In the report prepared by the Expert Panel, the *source zone* was defined as “the groundwater region (volume) in which DNAPL is present as a separate phase, either as randomly distributed sub-zones at residual saturations or ‘pools’ of accumulation above confining units and includes the volume of the aquifer that has had contact with free-phase DNAPL at one time, but where all of the DNAPL mass is now present only in the dissolved or sorbed phases or diffused into the matrix in fractured systems” (Kavanaugh et al., 2003). This article uses the same definition of a source zone. Note that even sites with relatively low concentrations can be included in this definition, as it includes sites where once-present NAPL is now present only in the dissolved or sorbed phase. While this article focuses on DNAPL source zones, many of the relationships can be applied to light nonaqueous phase liquid (LNAPL) source zones as well.

Source mass ( $M$ ) is defined as the total mass of contaminant in the source zone. The mass discharge rate ( $W$ ) is defined as the mass per time leaving the source zone. For this article, the flowrate through a source zone is assumed to stay constant as the source materials are removed, so that a change in the mass discharge rate is directly proportional to the change in average source concentration (Zhu & Sykes, 2004).

The amount of source depletion from active remediation (e.g., thermal, chemical oxidation, surfactants, biodegradation) is defined using a source-reduction factor labeled the remaining fraction (RF), or the fraction of total source mass remaining following remediation. The remediation time frame is defined as the time required for groundwater concentrations in the source zone to achieve some predetermined standard, such as maximum contaminant levels (MCLs).

While this article focuses on DNAPL source zones, many of the relationships can be applied to light nonaqueous phase liquid (LNAPL) source zones as well.

## MODEL DEVELOPMENT

Four simple concentration versus time models were used to evaluate the performance of source-depletion technologies (Exhibit 1):

- *Step Function Model*: Mass discharge rate (and average source concentration) remains constant as long as any source mass remains;
- *Linear Decay Model*: Mass discharge rate (and average source concentration) decreases linearly over time;
- *First-Order Decay Model*: Mass discharge rate (and average source concentration) follows a first-order decay pattern; and
- *Compound Step Function/First-Order Decay Model (“Compound Model”)*: Mass discharge rate (and average source concentration) remains constant until a certain fraction of the source mass is depleted, and then concentrations follow a first-order decay pattern.

For each model, planning-level equations are presented to show the RTF (1) under monitored natural attenuation (MNA) conditions and (2) after partial source depletion has been implemented. More importantly, the *relative* reduction in RTF due to partial source depletion is shown for each model. Design charts for estimating the change in the remediation time frame versus the amount of source depletion are presented for the four models.

### Step Function Model

For the step function model, the RTF for monitored natural attenuation is equal to the original source mass ( $M_0$ ) divided by the original mass discharge rate ( $W_0$ ) (sometimes referred to as the mass flux):

$$RTF_{MNA} = \frac{M_0}{W_0} \tag{1}$$

where  $RTF_{MNA}$  is the remediation time frame under MNA conditions (i.e., no source depletion),  $M_0$  is the original mass of contaminant in the source zone (NAPL phase, sorbed phase, dissolved phase, mass dissolved in matrix), and  $W_0$  is the original mass discharge rate from the source (mass per time).

The step function model assumes that the mass discharge rate remains constant as long as any source mass is present, so that the mass discharge rate does not change even after partial depletion of the source by remediation activities:

$$W(t) = W_0 \tag{2}$$

where  $W(t)$  = mass discharge rate at time  $t$  (mass per time).

The RTF for a source depletion project where the source mass is reduced to some RF value (i.e., an RF of 10 percent means only 10 percent of the original source mass remains, and that 90 percent of the mass has been removed by the project) can be calculated using the following expression:

$$RTF_{SD} = \frac{(M_0)(RF)}{W_0} \tag{3}$$

Design charts for estimating the change in the remediation time frame versus the amount of source depletion are presented for the four models.

**Exhibit 1.** Summary of source decay models

Source Model	Key Assumptions	$RTF_{MNA}$	$RTF_{SD}$	$\frac{RTF_{SD}}{RTF_{MNA}}$
Step Function	1. The mass discharge ( $W$ ) remains constant regardless of mass remaining in source ( $W_t = W_0$ )	$\frac{M_0}{W_0}$	$\frac{(M_0)(RF)}{W_0}$	$RF$
Linear Decay	1. The mass discharge rate decreases monotonically over time. 2. The slope of the mass discharge rate curve after source depletion is the same as the slope of the mass discharge rate curve before mass depletion. 3. The ratio of the cleanup goal mass discharge rate to the original mass discharge rate is much smaller than the source-reduction factor.	$\frac{2M_0}{W_0}$	$\frac{(2)(M_0)(RF)^{1/2}}{(W_0)}$	$RF^{1/2}$
First-Order Decay	1. The mass discharge rate decreases according to a first-order decay pattern. 2. The source mass is assumed to be proportional to the mass discharge rate at any time (e.g., if source depletion removes 50 percent of the mass, the mass discharge rate is reduced by 50 percent). 3. The source decay rate constant is the same before and after source depletion (i.e., the remaining mass after source depletion follows the pre-source-depletion pattern).	$\frac{-(M_0) \left[ \ln \left( \frac{W_g}{W_0} \right) \right]}{W_0}$	$-\left( \frac{M_0}{W_0} \right) \ln \left( \frac{W_g}{(W_0)(RF)} \right)$	$\frac{\ln \left( \frac{W_g}{(W_0)(RF)} \right)}{\ln \left( \frac{W_g}{W_0} \right)}$ or $\frac{\ln \left( \frac{C_g}{C_0 RF} \right)}{\ln \left( \frac{C_g}{C_0} \right)}$ (note 1)
Compound	1. Half of the source mass leaves the source zone as a constant mass discharge rate, and half leaves as with the mass discharge rate following a first-order decay process. Source-depletion case reduces the mass discharge rate by a factor equal to the source-depletion reduction factor.	$\left( \frac{(M_0)(0.5)}{(W_0)} \right) \left( 1 - \ln \left( \frac{W_g}{W_0} \right) \right)$	$\left( \frac{(M_0)(0.5)}{W_0} \right) \left( 1 - \ln \left( \frac{W_g}{(W_0)(RF)} \right) \right)$	$\frac{1 - \ln \left( \frac{W_g}{W_0 RF} \right)}{1 - \ln \left( \frac{W_g}{W_0} \right)}$ or $\frac{1 - \ln \left( \frac{C_g}{C_0 RF} \right)}{1 - \ln \left( \frac{C_g}{C_0} \right)}$ (note 1)

Note 1: Concentration-based equation can be used if the groundwater flowrate through the source zone does not change over time, and is not changed by the source-depletion project.

where  $RTF_{SD}$  is the remediation time frame for a source-depletion project, and  $RF$  equals the fraction of mass of contaminant remaining in the source zone immediately after the source-depletion project is completed (unitless).

Dividing Equation 3 by Equation 1 gives the fraction of the original remediation time frame that the source-depletion project with a given source mass RF will achieve:

$$\frac{RTF_{SD}}{RTF_{MNA}} = RF \text{ (for step function model)} \quad (4)$$

For example, if the step function model of source concentrations is applicable at a site, removing all but 10 percent of the source mass with some source-depletion technology (i.e., removing 90 percent of the mass or a  $RF = 0.10$ ) will reduce the site RTF to 10 percent of the RTF under MNA conditions (e.g., a 200-year remediation time frame will be reduced to a 20-year remediation time frame). Exhibit 2 presents a design chart summarizing the reduction in the remediation time frame relative to the source mass reduction factor.

### Linear Decay Model

The linear decay model assumes that the source zone mass discharge rate decreases monotonically over time, starting from an original source discharge rate. Therefore, the RTF for the MNA case is:

$$RTF_{MNA} = \frac{(2)(M_o)}{W_o} \quad (5)$$

For the case where source depletion is applied, an assumption is made that the slope of the mass discharge rate versus time curve is the same before and after source depletion, or:

$$\frac{W_o}{RTF_{MNA}} = \frac{W_{OSD}}{RTF_{SD}} \quad (6)$$

where  $W_{OSD}$  is the mass discharge rate immediately after source depletion (mass per time).

In addition, it is assumed that the mass discharge rate goal (i.e., the flowrate through the source zone times the concentration goal for site closure such as an MCL), when divided by the original mass discharge rate leaving the source zone, is much smaller than the RF associated with source depletion:

$$\frac{W_g}{W_o} \ll RF \quad (7)$$

where  $W_g$  is the mass discharge rate goal (i.e., the flowrate through the source zone times the concentration goal for site closure such as an MCL) (mass per time).

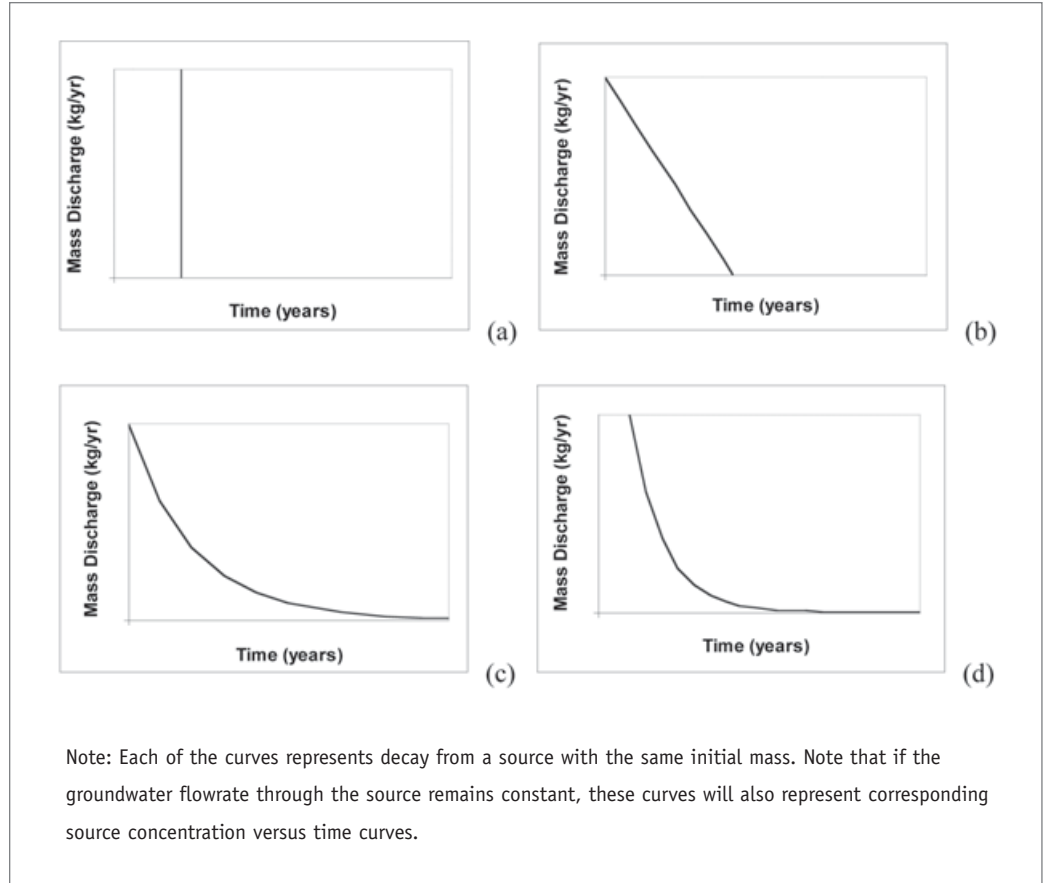
Therefore, the RTF for source depletion is:

$$RTF_{SD} = \frac{2(M_o)(RF)}{W_{OSD}} \quad (8)$$

Substituting Equation 6 yields:

$$(RTF_{SD})^2 = \frac{2(M_o)(RF)(RTF_{MNA})}{W_o} \quad (9)$$

The linear decay model assumes that the source zone mass discharge rate decreases monotonically over time, starting from an original source discharge rate.



**Exhibit 2.** Mass discharge rate versus time for (a) step function model, (b) linear decay model, (c) first-order decay model, and (d) compound model

Substituting Equation 5 yields:

$$RTF_{SD} = \frac{2(M_0)(RF)^{1/2}}{W_0} \tag{10}$$

Dividing Equation 10 by Equation 5 yields the fraction of the original remediation time frame that the source-depletion project with a given source mass RF will achieve:

$$\frac{RTF_{SD}}{RTF_{MNA}} = RF^{1/2} \tag{11}$$

In summary, the reduction in RTF for the linear decay model is proportional to the square root of the reduction factor (Exhibit 2).

### First-Order Decay Model

Several software systems have used a mass balance/mass flux approach for estimating plume response. Newell et al. (1996) used a simple box model and first-order source decay model in the BIOSCREEN model. This same box model was used for evaluating the effects of source decay in the BIOCHLOR model (Aziz et al., 2000) and the SourceDK decision support system (Farhat et al., 2004). A US EPA Issue Paper (Newell

et al., 2002) describes this model where concentrations versus time rate constants ( $k_{point}$ ) are used for estimating how quickly remediation goals will be met at a site.

The first-order decay model is based on the assumption that the mass flux from a naturally attenuating site follows a first-order decay pattern:

$$W_g = W_o \exp^{-(k_s)(RTF_{MNA})} \tag{12}$$

where  $k_s$  is the source decay coefficient (equivalent to  $k_{point}$  for concentration versus time data originating from the source zone). A mass balance shows that  $k_s$  is equal to the ratio of the mass discharge rate and the original source mass (Wiedemeier et al., 1999):

$$k_s = \frac{W_o}{M_o} \tag{13}$$

This relationship holds as long as the mass discharge rate at any time is assumed to be proportional to the source mass at any time:

$$\frac{M_o}{W_o} = \frac{M_t}{W_t} \tag{14a}$$

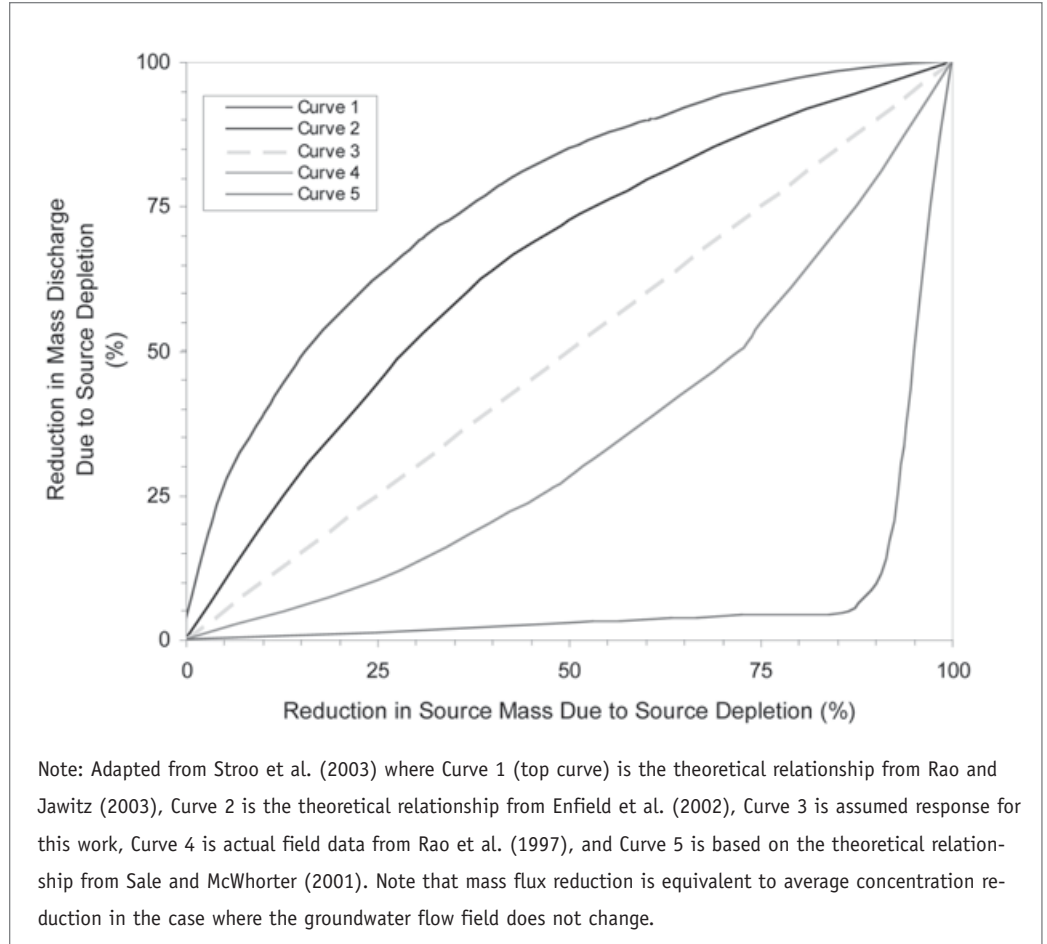
While quantifying the relationship between mass discharge rate and mass remaining in a source zone is a subject of continuing study (Falta et al., in press; Parker & Park, 2004; Zhu & Sykes, 2004), the assumption that the change in the mass discharge rate is directly proportional to the change in the source mass represents a “middle-of-the-road” case. Exhibit 3, adapted from Stroo et al. (2003), shows five mass flux response curves. The 45° line in the middle represents the case where mass flux reduction is directly proportional to source mass reduction (the assumption used in this work). Two theoretical curves are above the directly proportional line, and one theoretical and one actual field response curve are below the line. Determining the shape of the mass discharge response curve under MNA or source-depletion projects is difficult using today’s site characterization technology and current understanding of how source zone architecture affects mass discharge rate (Kavanaugh et al., 2003), meaning that a “middle-of-the-road” approach is appropriate for a planning-level model.

Data from a DNAPL remediation survey (Geosyntech Consultants, 2004) also support use of the directly proportional mass discharge/source mass relationship. Four of nine respondents indicated > 80 percent mass removals and > 80 percent reduction in mass discharge (approximately proportional response). Two respondents indicated > 80 percent mass removals and 60–80 percent reduction in mass discharge (lower-than-proportional response). One respondent indicated 50–80 percent mass removal and > 80 percent reduction in mass discharge, and two respondents showed 25–50 percent mass removal with 40–80 percent reduction in mass discharge (likely higher-than-proportional response).

Based on this middle-of-the-road approach and the mass balance relationships described in Equations 12 and 13, it can be assumed that the source decay constant ( $k_s$ ) is the same before and after source depletion. In other words, while source mass is removed by source depletion, the first-order decay characteristics of the remaining mass are assumed to be the same:

$$\frac{M_o}{W_o} = \frac{M_{OSD}}{W_{OSD}} \tag{14b}$$

Data from a DNAPL remediation survey also support use of the directly proportional mass discharge/source mass relationship.



**Exhibit 3.** Reduction in mass discharge (%) versus reduction in source mass (%)

The RF is the ratio of the source mass immediately after source depletion to the original mass:

$$M_{OSD} = (M_o)(RF) \quad (15)$$

By Equation 14b, the same holds true for mass discharge rate:

$$W_{OSD} = (W_o)(RF) \quad (16)$$

Rearranging Equation 12 and substituting Equation 13:

$$RTF_{MNA} = \frac{-(M_o) \left[ \ln \left( \frac{W_g}{W_o} \right) \right]}{W_o} \quad (17)$$

For the source-depletion case, rearranging Equation 12 and substituting Equations 13 and 15 yields:

$$RTF_{SD} = - \left( \frac{M_{OSD}}{W_{OSD}} \right) \ln \left( \frac{W_o}{W_{OSD}} \right) \quad (18)$$

Substituting Equation 14b yields:

$$RTF_{SD} = - \left( \frac{M_o}{W_o} \right) \ln \left( \frac{W_g}{W_{OSD}} \right) \quad (19)$$

Substituting Equation 16:

$$RTF_{SD} = - \left( \frac{M_o}{W_o} \right) \ln \left( \frac{W_g}{(W_o)(RF)} \right) \quad (20)$$

Dividing Equation 20 by Equation 17 and rearranging yields the fraction of the original RTF that the source-depletion project with a given source mass RF will achieve:

$$\frac{RTF_{SD}}{RTF_{MNA}} = \frac{\ln \left( \frac{W_g}{(W_o)(RF)} \right)}{\ln \left( \frac{W_g}{W_o} \right)} \quad (21)$$

In summary, the reduction in the RTF for the first-order decay model will be a function of the RF and the ratio of the mass discharge rate goal to the mass discharge rate before source-depletion efforts began. Note that the mass discharge rate is assumed to be directly proportional to the concentration, so that Equation 21 can be expressed in terms of concentration:

$$\frac{W_g}{W_o} = \frac{C_g}{C_o} \quad (22)$$

$$\frac{RTF_{SD}}{RTF_{MNA}} = \frac{\ln \left( \frac{C_g}{(C_o)(RF)} \right)}{\ln \left( \frac{C_g}{C_o} \right)} \quad (23)$$

where  $C_g$  is the concentration goal (such as an MCL) and  $C_o$  is the original concentration before source depletion began (mass per volume).

### Compound Model

A simplified form of the compound model is shown below, where it is assumed that half of the source mass leaves the source zone as a constant concentration and half leaves as a subsequent first-order decay process. During source depletion, the mass discharge rate is reduced by an amount equivalent to the reduction factor, and then remains at this rate until half the remaining mass is removed by natural attenuation. The derivation for the compound model is similar to the first-order decay model:

$$RTF_{MNA} = - \left( \frac{M_o}{W_o} \right)^{(0.5)} \left[ 1 - \ln \left( \frac{W_g}{W_o} \right) \right] \quad (24)$$

$$RTF_{SD} = - \left( \frac{M_o}{W_o} \right)^{(0.5)} \left[ 1 - \ln \left( \frac{W_g}{(W_o)(RF)} \right) \right] \quad (25)$$

As with the previous models, dividing the two equations results in an estimate of the fraction of the original RTF that the source-depletion project (with a given RF) will achieve:

During source depletion, the mass discharge rate is reduced by an amount equivalent to the reduction factor, and then remains at this rate until half the remaining mass is removed by natural attenuation.

$$\frac{RTF_{SD}}{RTF_{MNA}} = \frac{1 - \ln \left[ \frac{W_g}{W_0(RF)} \right]}{1 - \ln \left[ \frac{W_g}{W_0} \right]} \quad (26)$$

Using the previous assumption that concentration and mass discharge are proportional when the flow rate remains unchanged yields the concentration-based relationship:

$$\frac{RTF_{SD}}{RTF_{MNA}} = \frac{1 - \ln \left[ \frac{C_g}{C_0(RF)} \right]}{1 - \ln \left[ \frac{C_g}{C_0} \right]} \quad (26)$$

The current decision-making process for undertaking NAPL source depletion suffers from a lack of pertinent predictive models or other analytical tools to aid in providing reasonable estimates of performance.

## MODEL APPLICATION

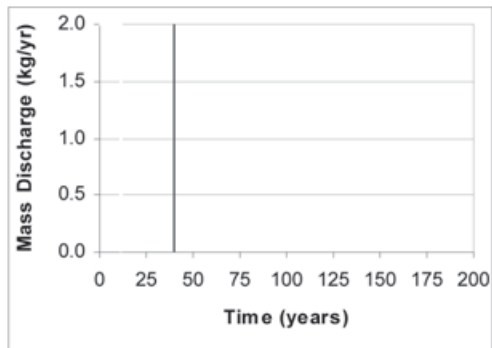
The current decision-making process for undertaking NAPL source depletion suffers from a lack of pertinent predictive models or other analytical tools to aid in providing reasonable estimates of performance. While source depletion will reduce source longevity, the benefit is difficult to predict because of uncertainty in source mass estimates and the distribution of the source mass after treatment (Kavanaugh et al., 2003). Models can serve as valuable guidance tools in measuring the impact of source zone depletion on the remediation time frame (Stroo, personal communication), particularly in the absence of long-term performance data from NAPL-contaminated sites (Stroo et al., 2003). The goal of this investigation was to develop planning-level calculations that could provide a basis for comparing various alternatives without being overly reliant on site-specific information that cannot be easily collected using current technologies.

The four models presented in this article range in complexity, but they all share a relatively simple framework for assessment. Specifically, they use the time required to remediate NAPL below a particular threshold as a metric. This value is referred to as the RTF and is quantified based on either (1) completely depleting the source mass (step function and linear decay models) or (2) depleting the source mass such that a goal concentration or mass discharge rate is achieved (first-order decay and compound models). While this value is of interest in terms of providing a discrete estimate of remediation time, it is more relevant to the decision to implement aggressive source treatment if it can be compared to the “no-go” alternative (MNA).

The finding that the lowest RTF values result from the step function and the highest result from the first-order decay model is illustrated in Exhibit 4. This example demonstrates the application of these planning-level calculations for each model in determining the benefit of source depletion relative to MNA. For each model, a case is considered where the initial mass of contaminant is 80 kg with the majority present as NAPL. The original mass discharge rate ( $W_0$ ) has been measured and determined to be 2 kg/yr.

### *Example of Step Function Model*

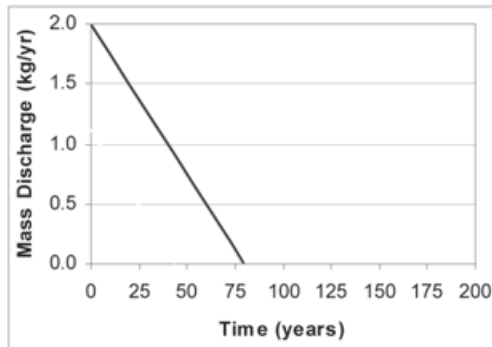
In the step function model, the mass discharge rate remains constant over time (Exhibit 4a). In the absence of a source-depletion strategy, the time required to remediate the



(a)

### Step Function

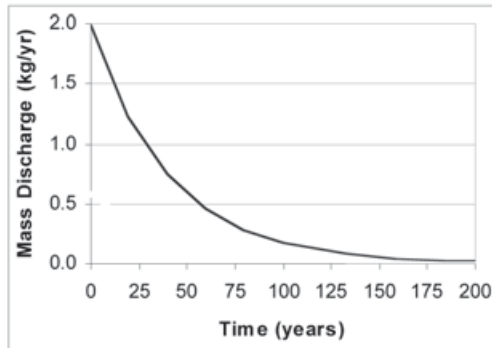
$$\begin{aligned}
 M_o &= 80 \text{ kg} \\
 W_o &= 2 \text{ kg/yr} \\
 \text{RF} &= 0.30 \text{ (70\% mass removed, 30\% remains)} \\
 \text{RTF}_{\text{MNA}} &= M_o/W_o = 80/2 = 40 \text{ years} \\
 \text{RTF}_{\text{SD}} &= (\text{RTF}_{\text{MNA}})(\text{RF}) = (40)(0.3) = 12 \text{ years}
 \end{aligned}$$



(b)

### Linear Decay

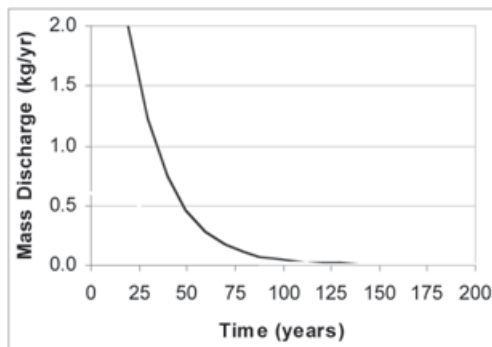
$$\begin{aligned}
 M_o &= 80 \text{ kg} \\
 W_o &= 2 \text{ kg/yr} \\
 \text{RF} &= 0.30 \text{ (70\% mass removed, 30\% remains)} \\
 \text{RTF}_{\text{MNA}} &= (2)(M_o)/W_o = (2)(80)/2 = 80 \text{ years} \\
 \text{RTF}_{\text{SD}} &= (\text{RTF}_{\text{MNA}})(\text{RF})^{1/2} \\
 &= (80)(0.3)^{1/2} = 44 \text{ years}
 \end{aligned}$$



(c)

### First-Order Decay

$$\begin{aligned}
 M_o &= 80 \text{ kg} \\
 W_o &= 2 \text{ kg/yr} \\
 W_g/W_o &= 0.01 \\
 \text{RF} &= 0.30 \text{ (70\% mass removed, 30\% remains)} \\
 \text{RTF}_{\text{MNA}} &= -(M_o/W_o)\ln(W_g/W_o) \\
 &= -(80/2)(\ln(0.01)) = 184 \text{ years} \\
 \text{RTF}_{\text{SD}} &= -(M_o/W_o)\ln(W_g/((\text{RF})(W_o))) \\
 &= -(80/2)\ln(0.01/0.3) = 136 \text{ years}
 \end{aligned}$$



(d)

### Compound

$$\begin{aligned}
 M_o &= 80 \text{ kg} \\
 W_o &= 2 \text{ kg/yr} \\
 W_g/W_o &= 0.01 \\
 \text{RF} &= 0.30 \text{ (70\% mass removed, 30\% remains)} \\
 \text{RTF}_{\text{MNA}} &= -(0.5(M_o/W_o))(1-\ln(W_g/W_o)) \\
 &= -(0.5(80/2))(1-\ln(0.01)) = 112 \text{ years} \\
 \text{RTF}_{\text{SD}} &= -(0.5(M_o/W_o))(1-\ln(W_g/W_o/\text{RF})) \\
 &= -(0.5(80/2))(1-\ln(0.01/0.3)) = 88 \text{ years}
 \end{aligned}$$

Note: Solid line represents mass discharge rate versus time for MNA case. Stippled line represents mass discharge rate vs. time for source-depletion case with source-reduction factor (RF) = 0.30 (i.e., 70 percent reduction in mass and 30 percent source mass remaining).

**Exhibit 4.** Examples of source decay models for case where  $M_o = 80 \text{ kg}$ ,  $W_o = 2 \text{ kg/yr}$ , and  $W_g/W_o = 0.01$

source mass due to natural attenuation ( $RTF_{MNA}$ ) is simply the mass divided by the discharge rate, which is equal to 40 years. If a source-depletion technology were implemented to remove 70 percent of the mass ( $RF = 0.30$ ), then the remaining 30 percent would flow out of the source zone at a rate of 2 kg/yr for 12 years until the entire mass was depleted. For an RF equal to 30 percent, this would require an  $RTF_{SD}$  of 12 years. Therefore, the step function model would predict a decrease in the RTF of 28 years as a result of choosing source depletion, a decrease of 70 percent that matches the decrease in the NAPL mass that was achieved by selecting an active remediation. Note that since the axes in Exhibit 4 are the mass discharge rate (y-axis) and time (x-axis), the area under each curve represents the mass that remains following source depletion (or the mass that is present originally in the case of natural attenuation).

The linear decay model requires that the mass discharge rate will decrease over time as the source mass is attenuated and flushed out of the source zone.

### *Example of Linear Decay Model*

The linear decay model (Exhibit 4b) requires that the mass discharge rate will decrease over time as the source mass is attenuated and flushed out of the source zone. The discharge rate reaches zero at the point in time when the source mass is entirely removed. In the example with an initial mass ( $M_0$ ) of 80 kg and an original discharge ( $W_0$ ) rate of 2 kg/yr, the natural attenuation alternative will require 80 years to remediate the entire contaminant mass. When source depletion is selected to remove 70 percent of the original mass, the remaining mass (24 kg) discharges at an initial rate ( $W_{OSD}$ ) that is less than  $W_0$  but decreases on a similar slope. The remediation time frame for this source-depletion alternative ( $RTF_{SD}$ ) can be calculated by applying Equation 11 to the case where  $RF = 0.3$  (or 30 percent). This yields a required time ( $RTF_{SD}$ ) of 44 years, which represents a 45 percent improvement over the MNA time frame. Note that  $W_{OSD}$  for this case is equal to 1.09 kg/yr.

### *Example of First-Order Decay Model*

In the first-order decay model (Exhibit 4c), the mass discharge rate also decreases over time in proportion to the mass present. Because this type of exponential decay will never result in a discharge rate equal to zero, a “goal” discharge rate (or concentration) can be preselected as the desired endpoint. For the example case with an initial mass ( $M_0$ ) of 80 kg and an original discharge ( $W_0$ ) rate of 2 kg/yr, a further assumption is made that a target discharge rate ( $W_g$ ) that is 1 percent of the original discharge rate will be sufficient to meet remediation goals. This would be equivalent to decreasing the concentration leaving the source zone from 0.5 mg/L to 0.005 mg/L. In a natural attenuation scenario, the time to reach this goal ( $RTF_{MNA}$ ) would be equal to 184 years. If 70 percent of this original mass were removed by using an aggressive source-depletion technology, then the resulting discharge rate ( $W_{OSD}$ ) would be smaller but decrease over time according to the same decay characteristics (identical  $k_s$  values). This yields an  $RTF_{SD}$  of 136 years, which represents a 26 percent improvement over the time frame for MNA.

### *Example of Compound Model*

When the compound model is applied (Exhibit 4d), the mass discharge rate remains constant until half of the mass leaves the source zone. The rate then declines expo-

nentially as the remaining mass is reduced to some target endpoint (such as  $W_g/W_0 = 0.01$ ). Therefore, an MNA strategy would result in half (40 kg) of the original 80 kg of contaminant leaving at a constant mass discharge rate of 2 kg/yr, a process that would take 20 years. The remaining 40 kg would leave at a declining discharge rate until the rate reaches 0.02 kg/yr. This goal is achieved after 92 years, yielding a total time frame under MNA ( $RTF_{MNA}$ ) of 112 years. A source-depletion technology that reduced the original mass by 70 percent would also decrease the original mass discharge rate by 70 percent (to 0.6 kg/yr). The remaining mass (after source depletion) would be 24 kg, of which 12 kg would flow out of the source zone at this constant rate. Following this 20-year period, an additional 68 years would be required before the final 12 kg of contaminant would be reduced to the targeted amount. This yields an  $RTF_{SD}$  of 88 years, which represents a 21 percent improvement over the time frame for MNA.

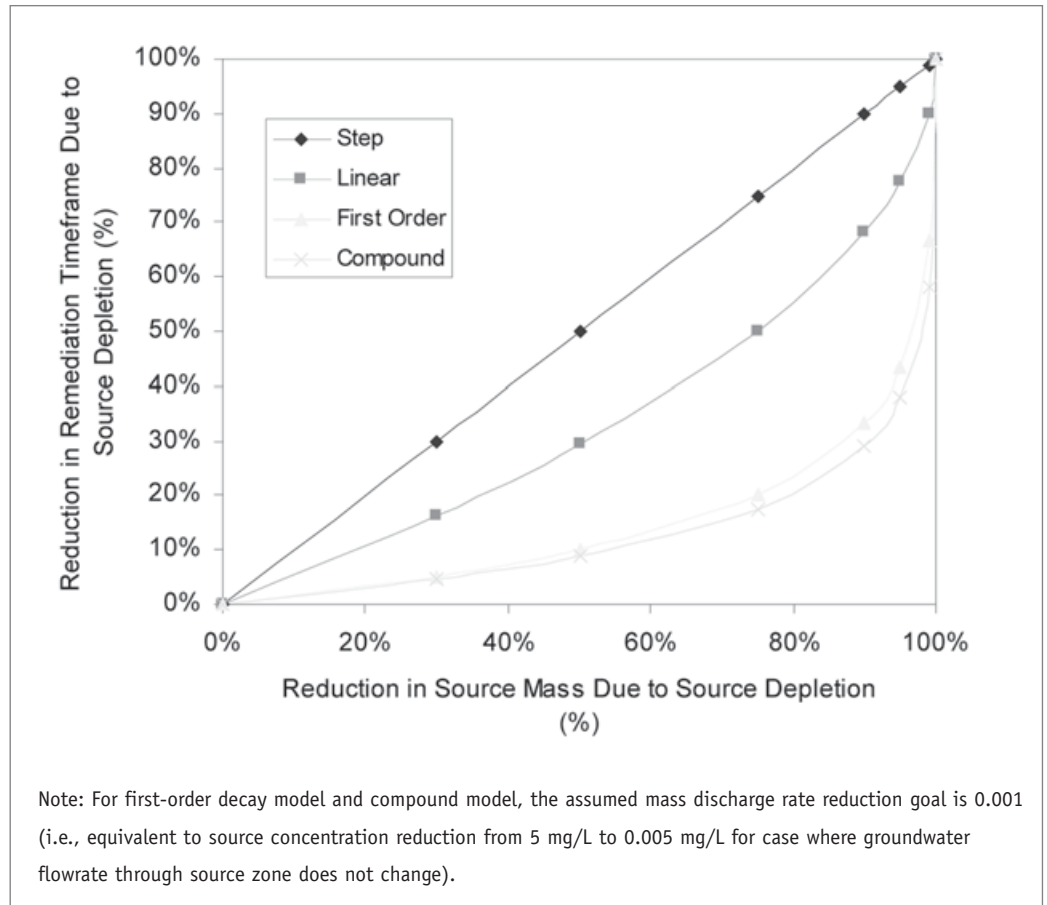
### Model Comparison

The preceding example demonstrates that the shortest RTF will always occur when the step function model is applied. Similarly, the next lowest RTF value for natural attenuation is achieved if the linear model is employed because the decrease in the mass discharge rate is not particularly severe. The RTF for the linear decay model will always be exactly twice the duration produced by the step function model. Both the compound and the first-order decay models rely on the assumption that a concentration (or mass discharge rate) of zero is a limit function, so each model must specify a goal concentration. If a conservative (low) goal is selected (such as one that meets MCLs for drinking water), then the remediation times must increase correspondingly. While the compound model will always predict a shorter RTF than the first-order decay model because the initial half of the mass is allowed to decrease at a constant rate, it is clear from the example that the total RTF in the compound model is dominated by the decay portion of the mass discharge period. In the MNA case where a modest reduction goal (0.01) was selected, discharge of the second 40 kg of contaminant mass represented 82 percent (88 of 112 years) of the total  $RTF_{MNA}$ .

Using these models to consider the benefit derived from source depletion generates a slightly different order of preference. The biggest improvement in the RTF for source depletion over MNA is observed for the step function model. This is a direct consequence of the fact that changes in the RTF for this model are proportional to changes in the RF. It can be imagined as the “equal benefit for effort” scenario following the decision to employ some type of source-depletion technology. In the preceding example, a 70 percent reduction in mass reduced the RTF by 70 percent as well (40 to 12 years). Applying source depletion for a linear decay model yielded an improvement in the RTF of 45 percent (80 to 44 years) relative to MNA. The first-order decay benefit achieved by source depletion was 26 percent (184 to 136 years), which is greater than the 21 percent benefit achieved when the compound model is employed. This degree of reduction in the RTF ( $(RTF_{MNA} - RTF_{SD})/RTF_{MNA}$ ) will always be greater for the first-order decay model than the compound model, regardless of the RF or the  $W_g/W_0$  goal selected.

A summary of this type of analysis is presented as a series of design curves in Exhibit 5. For each model type, the percent reduction in the RTF expected following the implementation of aggressive source depletion is shown. In this case, a reduction goal of 0.001 was selected, which is equivalent to decreasing the concentration leaving

The RTF for the linear decay model will always be exactly twice the duration produced by the step function model.



**Exhibit 5.** Relationship between source reduction factor and relative reduction in remediation time frame compared to MNA for four source decay models

the source zone from 5 mg/L to 0.005 mg/L (the MCL of TCE, for example). The exhibit illustrates the nonlinear relationship between RF and the reduction in the RTF in the three decay-based models. A similar set of design curves can be developed for any reduction goal. For stricter remedial objectives (lower ratios of  $C_g/C_o$ ), the first-order and compound models will deviate further from the step function and linear models, and thus predict diminishing reductions in the RTF.

### Most Representative Models

The models presented in this article follow an approach used to demonstrate a power-law relationship between source concentration as a function of source mass (Falta, 2003; Parker & Park, 2004; Rao & Jawitz, 2003; Zhu & Sykes, 2004):

$$\frac{C(t)}{C_o} = \left[ \frac{C_g}{C_o(RF)} \right]^\Gamma \tag{28}$$

where  $\Gamma$  is an empirical parameter. In this article, the step function model represents a  $\Gamma = 0$ , the linear decay model represents a  $\Gamma = 0.5$ , and the first-order decay model represents a  $\Gamma = 1.0$ . This relationship is used to demonstrate the impact of changes in

source strength (or concentration) on the remediation time frame following source depletion, and an assessment of the appropriateness of the various models.

The step function model represents the simplest case with the lowest RTFs and the greatest degree of improvement in RTFs following source depletion. It is similar to a conceptual model of DNAPL sites where substantial reductions in contaminant mass must be achieved before exiting concentrations begin to decrease (Sale & McWhorter, 2001).

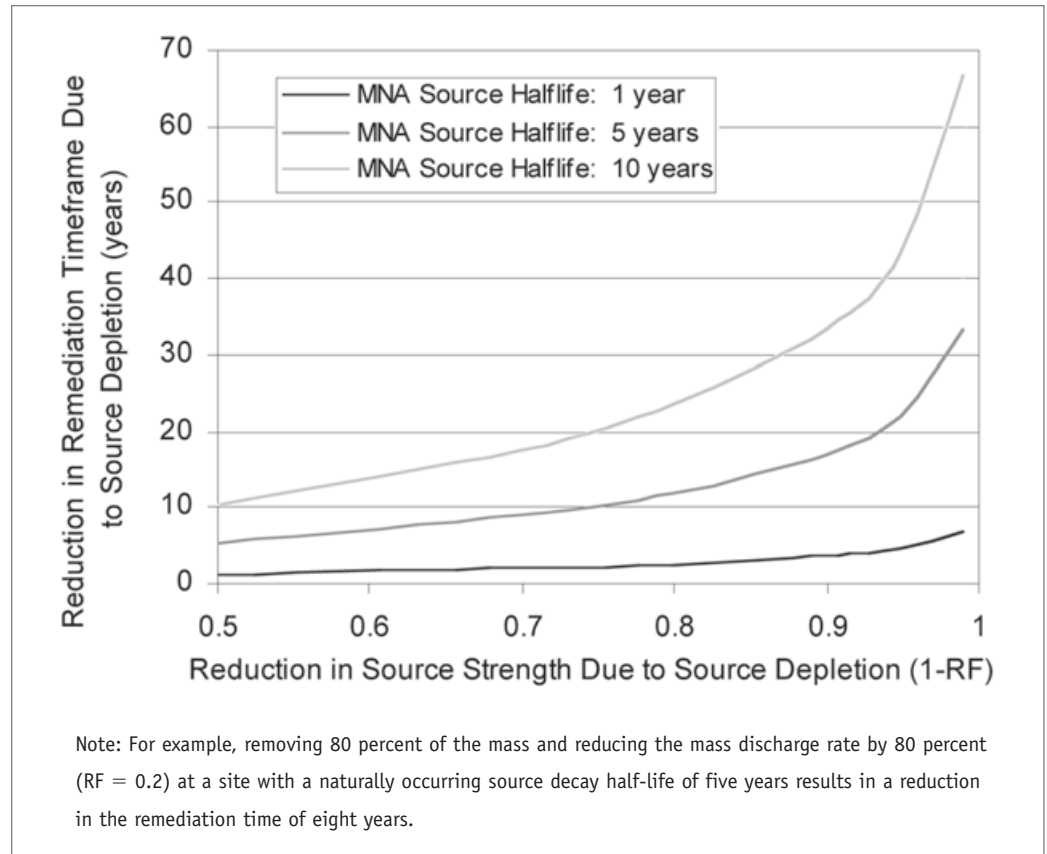
However, the fact that the mass discharge rate is independent of mass in this model is not a relationship that can be easily correlated with typical perceptions about common source zone architectures at actual DNAPL sites. For the source strength to remain constant over time, the surface area of a NAPL exposed to groundwater not at the effective solubility of the NAPL would need to remain relatively uniform even as mass was being depleted. This scenario could occur in cases where all NAPL is present in a large pool such that the NAPL surface area that forms an interface with the surrounding water matrix is minimized (Parker & Park, 2004). While this scenario is plausible in homogeneous and permeable aquifers with a large initial source mass, the mass discharge rates from these types of NAPL pools necessarily have lower initial mass discharge rates because of mass transfer limitations. A more likely conceptual model, applicable to most DNAPL sites, is that contaminant releases lead to extensive spreading of the DNAPL in a complex arrangement of thin, small DNAPL zones with relatively low DNAPL saturation (Gerhard & Kueper, 2003; McWhorter & Kueper, 1996; Zhu & Sykes, 2004).

Existing empirical evidence from chlorinated solvent sites suggests that the linear or first-order decay models are the most relevant in describing changes in the mass discharge rate. Laboratory-based studies such as those by Imhoff et al. (1994), Lamarche (1991), and Johnson & Pankow (1992) have demonstrated that flux rate and concentration decrease over time as source zones are depleted. A recent data compilation that analyzed long-term temporal trends (i.e., minimum record of five years and median record of nine years) from 45 groundwater-monitoring wells at 23 NAPL source zone sites concludes that most wells are characterized by decreasing concentrations over time (Newell et al., in press). At all of these sites, natural attenuation is the only means for source depletion. Both a linear regression model and a first-order decay model were statistically confirmed better fits for the data than a step function model. Using trichloroethylene as a typical DNAPL contaminant, the median first-order decay coefficients ( $k_{point}$ ) for 13 TCE sites were calculated to be 0.11 per year (half-life of 6.1 years). The  $k_{point}$  values for five 1,1,1-trichloroethane sites were even higher, with a median value of 0.41 per year (half-life of 1.7 years).

These data represent a shift away from a commonly accepted conceptual model of extremely long source zone contaminant concentrations over time. When viewed collectively, significant mass depletion appears to occur at these sites in the absence of aggressive source depletion within a period of time considerably shorter than the “decades or centuries” that has been previously cited (e.g., Feenstra et al., 1996; Kavanaugh et al., 2003). While this finding argues positively for the attenuation of source zones within shorter time frames, it also emphasizes that models that predict decreasing mass discharge rates over time (such as the linear, first-order, and compound models) are more appropriate than the step function model for predicting these time frames.

Process knowledge provides an additional basis for choosing the decay-based models as the most appropriate. The change in a DNAPL source zone over time can be described as a series of different attenuation processes superimposed on each other.

Existing empirical evidence from chlorinated solvent sites suggests that the linear or first-order decay models are the most relevant in describing changes in the mass discharge rate.



**Exhibit 6.** Reduction in remediation time frame (in years) versus reduction in source strength (1-RF) (source mass and mass discharge rate) due to source depletion for first-order decay source model and different MNA source decay half-lives.

DNAPL fingers (i.e., thin, vertically oriented DNAPL zones) will dissolve more quickly than DNAPL pools as the fingers have a higher surface area-to-volume ratio. In general, DNAPL pools with a short length in the direction of flow will dissolve faster than larger pools with a longer length in the direction of groundwater flow. DNAPL in zones with no or little groundwater flow will persist longer than zones with high groundwater flow. Matrix diffusion, linear desorption, desorption from the fraction with different equilibrium kinetics (Chen et al., 2002), and dispersion will all contribute to lower mass flux versus time at DNAPL sites. The combination of all of these processes indicates that some type of tailing is likely to be observed at DNAPL sites (Zhu & Sykes, 2004). The first-order and compound models are likely to best represent this tailing.

### Application to RTF Estimates

The planning-level approach detailed in this article provides a framework for addressing many of the key questions noted by the US EPA Expert Panel on DNAPL Remediation. Specifically, this is a simple tool to assess performance after the implementation of a source-depletion technology. While it does not provide guidance for predicting the performance (efficiency) of a source-depletion technology, the degree of source removal

**Exhibit 7.** Required input data to estimate relative reduction in RTF achieved by source depletion (relative to MNA)

Input Parameter	Used for Which Model?	Where Obtain Data?	Reported Values	Reference
Remaining Mass Fraction ( $RF$ )	<ul style="list-style-type: none"> <li>• Step Function (Eq. 4)</li> <li>• Linear Decay (Eq. 11)</li> <li>• First-Order Decay (Eqs. 21, 23)</li> <li>• Compound (Eqs. 26, 27)</li> </ul>	Performance data for source-depletion projects	0.01 to 0.5 (median from 59 sites: 0.12)	McGuire et al., in press
Remediation Goal ( $W_g/W_0$ )	<ul style="list-style-type: none"> <li>• First-Order Decay (Eqs. 21, 23)</li> <li>• Compound (Eqs. 26, 27)</li> </ul>	Site data; divide maximum source concentration by conc. goal (e.g., MCL)	0.1 to 0.00001 (Typical source concentrations: 0.05 to 500 mg/L; typical remediation goals: 0.2 mg/L to 0.005 mg/L)	Newell et al., in press

(1- $RF$ ) is an input value that can be used to predict the benefit gained (in terms of the reduction in the remediation time frame) by implementing aggressive source depletion. Therefore, the planning-level tools in this article are intended to improve the decision-making process regarding whether to proceed with source depletion or to use a long-term containment or MNA approach.

To illustrate this point, the database-derived DNAPL half-lives of approximately two to ten years observed at field sites (Newell et al., in press) can be used to generate the reductions in RTF achieved by source reduction. This is illustrated in Exhibit 6 for cases where the half-life is set at one year, five years, and ten years (using the linear decay model with the assumption that the source decay coefficient  $k_s$  does not vary with time). By selecting a desired (goal) mass reduction from applying aggressive source depletion, the reduction in RTF (relative to MNA) can be determined for each case. For example, if 70 percent source removal were achieved by thermal treatment ( $RF = 0.30$ ), the “savings” in the RTF would range from 1.7 to 17 years, depending on the half-life. Using the median MNA source half-life of five years, the result is an improvement of 8.7 years. If a more optimal 90 percent source removal were achieved, then the savings in RTF would range from 3.3 to 33 years, with a median value of 17 years.

This quantitative assessment of the reduction in the RTF can be used to develop net-present value estimates of source depletion versus long-term containment, as discussed by Kavanaugh et al. (2003). Because this approach can incorporate the overall cost advantages (or disadvantages) of a reduction in the RTF in a net present value analysis, it provides a rationale and cost-driven framework for assessment.

Additional detail on the input data required to apply the predictive equations is presented in Exhibit 7 (for the relative reduction in RTF due to source depletion) and Exhibits 7 and 8 (for the absolute reduction in RTF due to source depletion).

The example described earlier in this article (Exhibit 4) demonstrates that a source-depletion project that achieved 70 percent mass removal reduced the RTF

**Exhibit 8.** Additional input data to estimate absolute reduction in RTF achieved by source depletion

Input Parameter	Used for Which Model?	Where Obtain Data?	Reported Values	Reference
Initial Mass Discharge Rate ( $W_0$ )	<ul style="list-style-type: none"> <li>• Step Function (Eqs. 1, 3)</li> <li>• Linear Decay (Eqs. 5, 10)</li> <li>• First-Order Decay (Eqs. 17, 20)</li> <li>• Compound (Eqs. 24, 25)</li> </ul>	Site monitoring data; modeling studies	0.3 to 425 g/day	Einarson & MacKay, 2001
Source Mass ( $M_0$ )	<ul style="list-style-type: none"> <li>• Step Function (Eqs. 1, 3)</li> <li>• Linear Decay (Eqs. 5, 10)</li> <li>• First-Order Decay (Eqs. 17, 20)</li> <li>• Compound (Eqs. 24, 25)</li> </ul>	Site sampling data	—	Kavanaugh et al., 2003

from 184 to 136 years (26 percent) when a first-order decay model was applied and a conservative concentration-endpoint ( $C_g/C_0 = 0.01$ ) was selected. While a comprehensive cost analysis was not conducted for this scenario, the relatively minor improvement in the RTF might make it difficult to favor the selection of an aggressive source-depletion strategy based on reduction in RTF alone. Other benefits of source depletion related to depletion of contaminant mass that could be incorporated into a life-cycle cost analysis include: (1) mitigating the future potential for human contact and exposure through long-term reduction of volume, toxicity, and mobility of the DNAPL; (2) mitigating the future potential for unacceptable ecological impacts; (3) minimizing risks of failure of long-term containment strategies; (4) mitigating public stakeholders' concerns; (5) enhancing a company's "green image" as stewards of the environment; and (6) minimizing future uncertain transaction costs associated with management of the site (Kavanaugh et al., 2003). The tools described in this article do not explicitly address these other benefits.

In summary, the intention of this work is to provide a planning-level quantitative context for assessing the benefits of source depletion. The current tools can be used to generate:

- *absolute* RTF estimates (in years) =  $f$ (current mass discharge rate, current source mass, remediation goal, and source reduction factor)
- *relative* RTF estimates (as fraction of MNA RTF) =  $f$ (remediation goal and source reduction factor only)

While existing DNAPL source zone models are available, in general they cannot be applied to specific sites because of the complexity of the input parameters (source zone architecture; location of source zone features such as pools, fingers, etc.; location of geologic heterogeneities; and mass transfer rates) and the uncertainties in simulating complex, interacting processes (*in situ* biodegradation, mass transfer rates, matrix diffusion). The anticipated use of the simple planning models will likely be to quantify the relative relationship between RTF and source depletion using commonly available site data.

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