

CONTAMINANT TRANSPORT IN GRANULAR MEDIA: A CASE STUDY IN COMPUTATIONAL MODELING

Dudley J. Benton, Ph.D.

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Abstract: Accurate modeling of contaminant transport is essential to effective containment design. Anything less may lead to failure, the consequences of which depend on the virulence and quantity of the contaminant. The mathematical model presented herein arose from such a failure. In spite of careful implementation, the impounded substance breached the barrier and escaped into the environment. An inadequate model used in the design phase overestimated the time to breach the cap by orders of magnitude. A more accurate model, correcting the original mistakes and accounting for the granularity of the media, yields a breach time consistent with the tragic failure.

Keywords: contaminant transport; granular media; barrier failure; diffusion; dispersion; advection; sorption

1. Introduction

Effective remediation strategies are essential to protecting the environment and its inhabitants. Ineffective strategies that lead to failure may result in greater harm than inaction because implementation can induce complacency and misplaced assurance of protection. It is perhaps more important to study failures than successes in the field of remediation so that they not be repeated. It is particularly instructive to investigate those failures whose cause has been ascertained. Breach of containment in the case of a carcinogen may exact a staggering toll. Any opportunity to learn from such a loss must not be wasted.

Failure of a barrier cap to contain the contaminant in-situ triggered an investigation. When that investigation proved unfruitful, this author was brought in as an uninvolved party to assess the data. A review of the model used to design the barrier quickly revealed two issues: one computational and a second related to material properties. Samples of the barrier material were collected after failure of the cap. These were sent to a laboratory for routine analysis. While not specifically requested, the laboratory provided a grain-size distribution, which is common for soil samples. The barrier material was clearly not amorphous, but this was assumed in the original model on which the design was based. These modeling flaws combined to over-estimate the effectiveness of the barrier and time to breach by orders of magnitude. Failure of the computational simulation of pollution dispersion in the design phase makes this a poignant case study.

2. Governing Equation and Basic Solutions

Contaminant transport in groundwater is often introduced as a one-dimensional partial differential equation including three components: retardation, dispersion and/or diffusion, plus advection. These represent temporal, concentration driven, and flowing processes.

$$R \frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial x^2} - V \frac{\partial C}{\partial x} \quad (1)$$

Here, C is the concentration, x is the spatial dimension, t is time, R is the advective retardation factor, D is the diffusion coefficient, and V is velocity (generally assumed to be of the aqueous phase). Separate advective and diffusive retardation factors are sometimes included in this equation. There is considerable discussion in the literature concerning the appropriate choice of values for R and D . This discussion will not be repeated here, as this is not the current focus. The paper by Bredehoeft and Pinder [6] is the seminal reference on this subject. Delle Site [9] is perhaps the most extensive collection of related properties for organic compounds. These two references also include several analytical solutions to Equation 1. Ogata and Banks [13] provide the following solution for a semi-infinite domain with initial concentration, C_0 :

$$C(x,t) = \left(\frac{C_0}{2} \right) \left[\operatorname{erfc} \left(\frac{x - \frac{Vt}{R}}{2\sqrt{Dt}} \right) + \exp \left(\frac{Vx}{D} \right) \operatorname{erfc} \left(\frac{x + \frac{Vt}{R}}{2\sqrt{Dt}} \right) \right] \quad (2)$$

Solutions for different boundary conditions may be found in several of the references, including van Genuchten and Alves [17]. Mohsen and Baluch [11] discuss solutions for various finite domains, while Jaiswal, Kumar Yadav [10] discuss temporal aspects of these same solutions. For the case of a uniformly contaminated slab of thickness, L , the solution becomes:

$$C(x,t) = \left(\frac{C_0}{2} \right) \left[\operatorname{erfc} \left(\frac{x - \frac{Vt}{R_A} - \frac{L}{2}}{2\sqrt{\frac{Dt}{R_D}}} \right) - \operatorname{erfc} \left(\frac{x - \frac{Vt}{R_A} + \frac{L}{2}}{2\sqrt{\frac{Dt}{R_D}}} \right) \right] \quad (3)$$

Results for typical properties ($C_0=1$; $L=0.5$ m; $R=60,000$; $V=3.3$ cm/day; $D=31.6$ cm²/yr) are shown in Figure 1. The solid curve (25 years) shows non-trivial movement of the contaminant over a distance of 0.5 m, which is within an order of magnitude of the case considered here. In spite of using the same properties, calculations employed in designing the failed barrier predicted far less movement of the contaminant over a much longer time span.

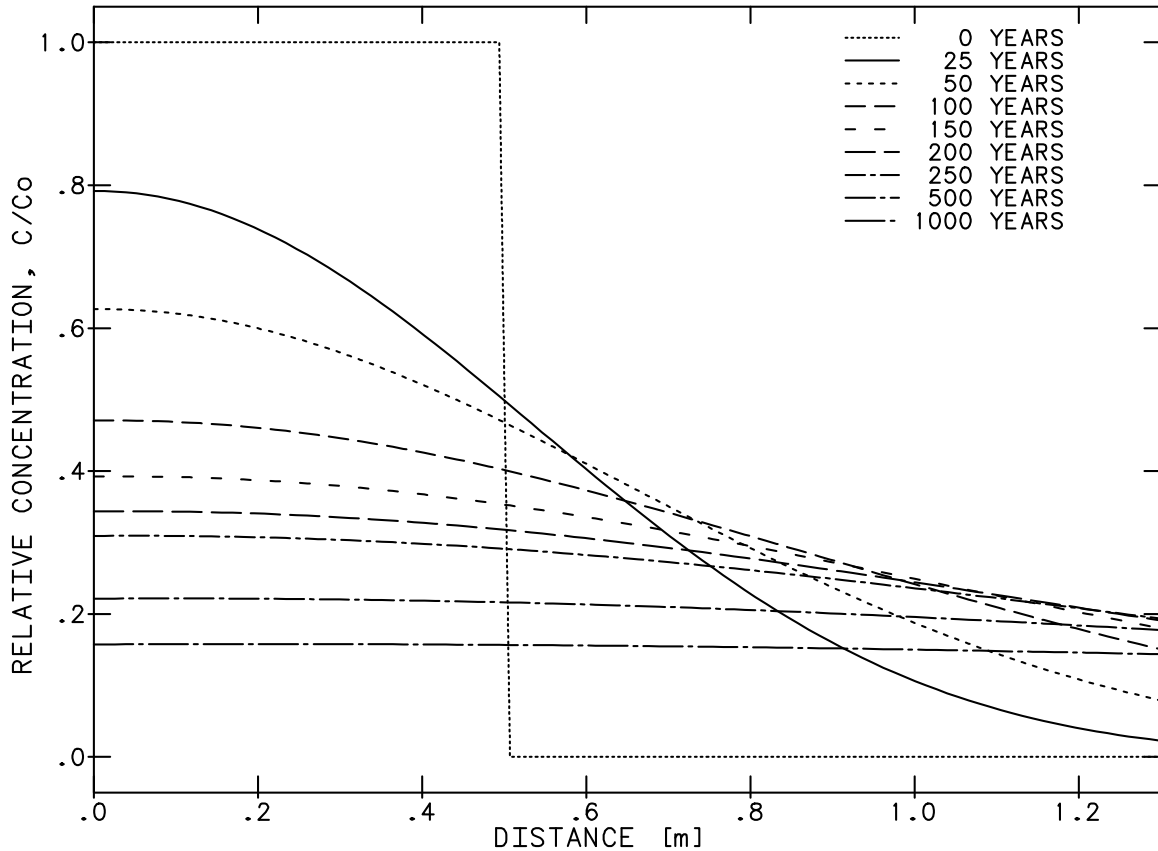


Figure 1. Typical Solutions to Equation 3

3. Computational Error in the Cap Design

The developer of the original model on which the failed design was based noted the source of the analytical solution used (the landmark paper by Bredehoeft and Pinder [6]). This solution is an algebraic variant of Equation 3, yielding the same concentrations values when supplied with equivalent properties, dimensions, and time. The predicted breakthrough time, however, was vastly greater than the preceding curves would seem to indicate and was borne out in experience. This discrepancy arose from two errors alluded to previously: mathematical and physical.

The Gauss error function, while unfamiliar to some, has been available in Excel® for a decade. However, it is not a native function in any programming language (e.g., FORTRAN, BASIC, Pascal, or C). This omission led to the first (computational) flaw in the cap design. The software used by the remediation contractor to design the barrier was written in FORTRAN. The source of the error function used in that program was also listed: CRC Standard Math Tables [5].

There is no simple formula for the error function; however, there are many sufficiently accurate approximations. The one most commonly used comes from Abramowitz and Stegun [1]. While the error function appears in several sections of the CRC Math Tables (e.g., Laplace

transforms), this reference does not contain an approximation. What it does contain is the Taylor series expansion:

$$\operatorname{erfc}(x) = 1 - \frac{2}{\sqrt{\pi}} \left(x - \frac{x^3}{3} + \frac{x^5}{5 \cdot 2!} - \frac{x^7}{7 \cdot 3!} + \dots \right) \quad (4)$$

The developer of the original FORTRAN model used only the first eight terms of this infinite series, which was a most unfortunate—and completely unnecessary—simplification. The asymptotic behavior of the error function is of primary interest in transport calculations and predicting breakthrough in particular. This fundamental behavior (or shape) was completely lost by truncating the series, as illustrated in Figure 2.

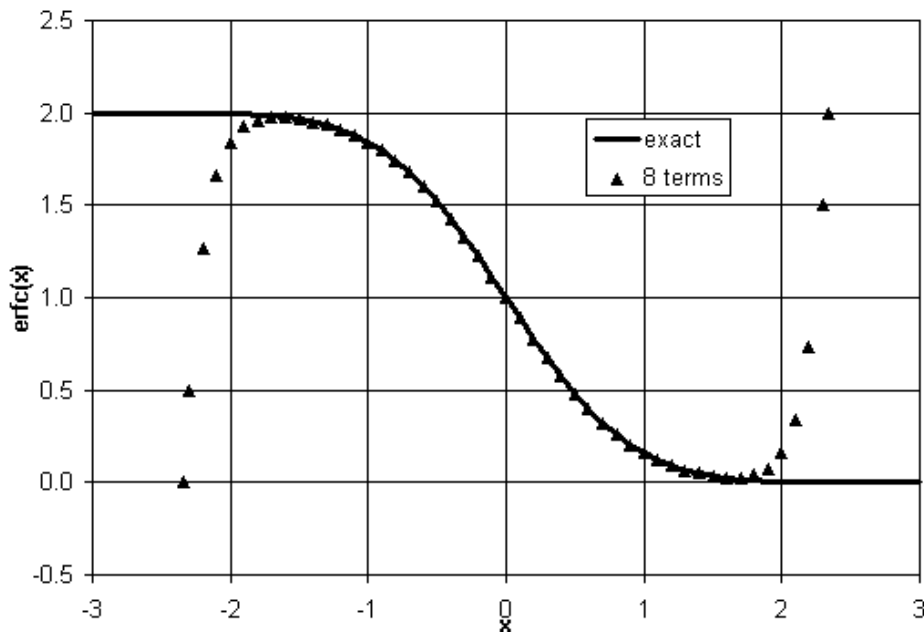


Figure 2. Error Function (Exact and Truncated)

There is no "arc" error function from which to back out time, t , from a particular location, x , so the FORTRAN program simply ran out values of time, looking for a positive concentration ($C > 0$) at the outer edge of the cap. The thickness of the cap was increased in steps until the breakthrough time exceeded the remediation requirement. The final result was in excess of 1000 years, considered more than adequate... until the interred carcinogen emerged in a recreation area.

4. Property Error in the Cap Design

The retardation factor is an empirical correction of a large-scale phenomenon (advection) in order to account for a small-scale phenomenon (sorption). An analogy would be a correction factor applied to the speed of a hiker stumbling through a briar patch. More receptor sites (thorns)

provide more retardation. When no more receptor sites are available, no more retardation can occur. This is borne out by the observation that even small amounts of Free Organic Carbon (FOC) will greatly increase the retardation of Polychlorinated Biphenyls (PCBs) (see discussion in Pankow and Cherry [15]). Guidance for estimating sorption parameters may be found in Piwoni and Keeley [16]. Moyo, Tandlich, Wilhemli, and Blatz [12] provide extensive discussion and references for organic compounds and their interaction with fine soils, such as was used in the cap under investigation.

Experimental verification of retardation factors is difficult to obtain, owing to the potentially long time scales. Several methods for improving the accuracy and contracting the process have been proposed, but these require considerable care in performance and application. Aniszewski [3] provides an excellent discussion of the methods as well as a test site that has proven a most useful evaluation opportunity. Further details of transport from landfill sites may be found in Varank, Demir, Sekman, and Bilgili [19] and Chen, Tu, Chen, and Chen [7].

This retardation factor should be applied directly to the aqueous advective transport mechanism. A different retardation factor for the host substance should be applied to the dissolved advective transport mechanism, as these two depend on differing species interactions. Use of the same retardation factor for both transport mechanisms would be inappropriate. Pankow and Cherry report much lower retardation factors for less hydrophobic organic compounds.

It is inappropriate to apply a retardation factor to diffusion. Still, the preceding transport model can be modified to account for retardation in several ways. A separate advective and diffusive retardation factor with the diffusive retardation factor much lower than the advective would be one modification to consider. Another possibility would be to model the media as a composite of spherical grains, considering only diffusion within the grains and diffusion plus advection between the grains, as discussed by Wu and Gschwend [20,21].

Advection of the contaminant in non-aqueous form, both non-aqueous advection and dissolved non-aqueous advection, may also be subject to retardation. Applying the same retardation factor to non-aqueous advection and aqueous advection would result in the two very different processes being mathematically indistinguishable. If this were done, the impact of solubility would be completely eliminated. The molecular contact between the surface of micro-droplets and the media is vastly less than that of the aqueous phase, which is in intimate contact with the media. It stands to reason that retardation, which depends on receptor sites, may be much different for contaminants in the aqueous phase than the non-aqueous, even though both are being transported by the same groundwater flow.

Replacing the truncated error function with the accurate approximation from Abramowitz and Stegun changed the result, but still didn't match the observed breakthrough time within two orders of magnitude. Based on a review of the aforementioned literature, reasonable property values were used in designing the barrier, eliminating this as the source of such a large

discrepancy. Property values were consistent with Anderson and Pankow [2], Palermo, Maynard, Miller, and Reible [14], and Wu and Gschwend [20,21]; however, Equation 5 was used in the original FORTRAN code for retardation factor.

$$R = 1 + \frac{\rho K}{\varepsilon} \quad (5)$$

where ρ is the density, K is the partitioning coefficient, and ε is the porosity. The preceding equation is incorrect. Equation 6 is the proper relationship (see Pankow and Cherry, Equation 8.2, p. 240):

$$R = \varepsilon + \rho K \quad (6)$$

It is evident from Equation 6 that partitioning coefficients are central to determining the time scale of advective and dispersive transport. These are dependent on the chemical species, the media, and the groundwater (or other host). Coefficients span orders of magnitude, as do the resulting retardation factors. Delle Site [9] provides a massive collection of these coefficients as well as an extensive discussion of their use and the impact these have on modeling results.

In his dissertation, Vandenbohede [18] focuses on the selection and estimation of modeling parameters for site characterization. He also provides an excellent discussion of dispersion vs. diffusion. Several computer models are compared in this investigation, along with the merits and strengths of each. The figures and graphs in this reference are particularly instructive, as are the two detailed case studies.

The porosity of the barrier material is typically small, especially for materials suitable for capping contaminants in-situ. Using Equation 5 over-estimates the retardation factor, erroneously increasing the breakthrough time. While correcting this error made a significant difference, it still yielded a breakthrough time much greater than the observed, indicating these two errors were not the sole deficiency of the original model.

5. Multiple Transport Mechanisms

Aniszewski [3] presents three significant processes considered in a large-scale transport experiment: advection, dispersion, and sorption. Aniszewski makes a strong case that none of these should be ignored when considering migration of hazardous substances in the environment. While it is not the same sort of flaw as the first two errors, failure to consider all of these mechanisms contributed to the inadequate barrier design under investigation.

Das, Sultana, and Singh [8] provide a thorough discussion of the Equation 1 as well as several analytical solutions, including: dispersion-dominated and advection-dominated cases. They also present variants of Equations 2 and 3, which may be used to approximate these limiting cases. Physical properties of the contaminant and media in the present case lie between these two

extremes so that neither process can be ignored. In their equations 20 and 21a through 21d, Das, Sultana, and Singh [8] present an essential step to arriving at a practical analytical solution more representative of the case under investigation. Specifically, they split the respective transport mechanisms into separate terms. The resulting transport mechanisms include:

1. Aqueous Diffusion (AD) (limited by the solubility in water)
2. Aqueous Advection (AA) (limited by the solubility in water)
3. Non-aqueous Advection (NA) (not limited by solubility)
4. Dissolved Advection (DA) in a host hydrocarbon (limited by the solubility in the host and by the solubility of the host in water)
5. Dissolved Non-aqueous advection (DN) in a host hydrocarbon (limited by the solubility in the host)

The first two transport mechanisms, aqueous diffusion (AD) and aqueous advection (AA), are limited by the solubility of the contaminant in water, which, in this case, is between 2.5 and 3.0 ppm. Samples collected at the site found pore water concentration to be approximately 9.0 ppm, indicating super-saturation. Total concentrations were found as high as 100 ppm, indicating bulk migration of the contaminant through the barrier. These values revealed that most of the contaminant was present in states other than that purely dissolved in the pore water. The third mechanism, non-aqueous advection (NA), is not limited by solubility. The fourth and fifth mechanisms, dissolved advection (DA) and dissolved non-aqueous advection (DN), are limited by the solubility in the host hydrocarbon, which is much higher in this case.

No more than two of the last three mechanisms may be considered insignificant, as this would leave no mechanism by which to explain presence of the contaminant in concentrations exceeding the solubility in water. The contaminant must have migrated to a sampling point by one or more of these mechanisms. If the non-aqueous phase transport mechanisms are lumped into the corresponding aqueous phase transport mechanisms, this effectively eliminates the impact of the solubility of the contaminant and host in water. If the dissolved phase transport mechanisms are lumped into the pure aqueous phase transport mechanisms, this effectively eliminates the impact of the solubility of the contaminant in the host. The computed total concentration of contaminant at the point of breakthrough is, therefore, highly dependent on which of these transport mechanisms are ignored and which are lumped together.

Diffusion is a molecular-scale phenomenon. Aqueous advection is a larger scale phenomenon, where the water and contaminant molecules behave as a fluid continuum. Non-aqueous advection occurs on a yet larger scale, where micro-droplets are discontinuities in the fluid continuum. Caution must be exercised when applying any mathematical relationship (i.e., Fick's Law of Diffusion), which was developed presuming a continuum, to a phenomenon involving discontinuities.

These minimal transport mechanisms, plus a necessity of retaining the distinction between saturation-limited and super-saturation pathways, determine which parts of the aforementioned composite solution of Das, Sultana, and Singh are essential for adequately representing the processes in the present case. The contaminant may be seen as an initial source in contact with an uncontaminated semi-infinite domain—the barrier and ultimately the environment. Equation 7 is the simplest solution to this problem that meets these requirements.

$$C(x,t) = \left(\frac{C_S}{2} \right) \left[\operatorname{erfc} \left(\frac{x - \frac{Vt}{R_{AA}} - \frac{L}{2}}{2 \sqrt{\frac{D_A t}{R_{AD}}}} \right) - \operatorname{erfc} \left(\frac{x - \frac{Vt}{R_{AA}} + \frac{L}{2}}{2 \sqrt{\frac{D_A t}{R_{AD}}}} \right) \right] + \frac{(C_0 - C_S)}{2} \left[\operatorname{erfc} \left(\frac{x - \frac{Vt}{R_{DA}} - \frac{L}{2}}{2 \sqrt{\frac{D_D t}{R_{DN}}}} \right) - \operatorname{erfc} \left(\frac{x - \frac{Vt}{R_{DA}} + \frac{L}{2}}{2 \sqrt{\frac{D_D t}{R_{DN}}}} \right) \right] \quad (7)$$

In Equation 7, C_0 is the initial concentration, C_S is the solubility, and L is the thickness of the cap. Equations 2 and 3 ignore the solubility, blurring the transport mechanisms; whereas, Equation 7 separates these two and accounts for solubility. This more general equation also includes two separate diffusion coefficients (cap material and pore water) as well as four different retardation factors (associated with four of the five different modes of transport mentioned previously and designated: AA, AD, DA, and DN). This solution may be adequate for a continuous (amorphous) media, but it does not account for finite grain size.

6. Granular Media

The original model used to design the cap assumed a continuous (amorphous) media. Few capping materials besides plastic would fit this description. The grain size distribution analysis provided by the soils laboratory revealed the barrier was, in fact, granular. Rather than being continuous, this domain consists of grains plus pore water. The grains are of non-uniform size, but will be considered to have uniform composition (i.e., physical properties). Wu and Gschwend [20,21] approximated transient diffusion within a single spherical grain of radius, r , by replacing $x - Vt/R - L/2$ with $r - d/2$ in the preceding solution for a semi-infinite slab. There is no pore water within a grain, which eliminates the velocity term from the intra-grain solution.

It remains to characterize the granularity of the media and incorporate this into the calculations. This characterization might be done in several ways, the most obvious being the standard sizes used to classify the samples. This choice proved adequate, plus it is naturally extended to other cases, as soil analyses are reported in these gradations. Figure 3 shows the grain

size distribution based on eight samples taken from the cap and tested by the soils laboratory. A straight line on this log-probability graph corresponds to a lognormal distribution. The data fit such a line quite well. In this case, the mean is $\mu=10^{-0.53}$ and standard deviation $\sigma=0.79$.

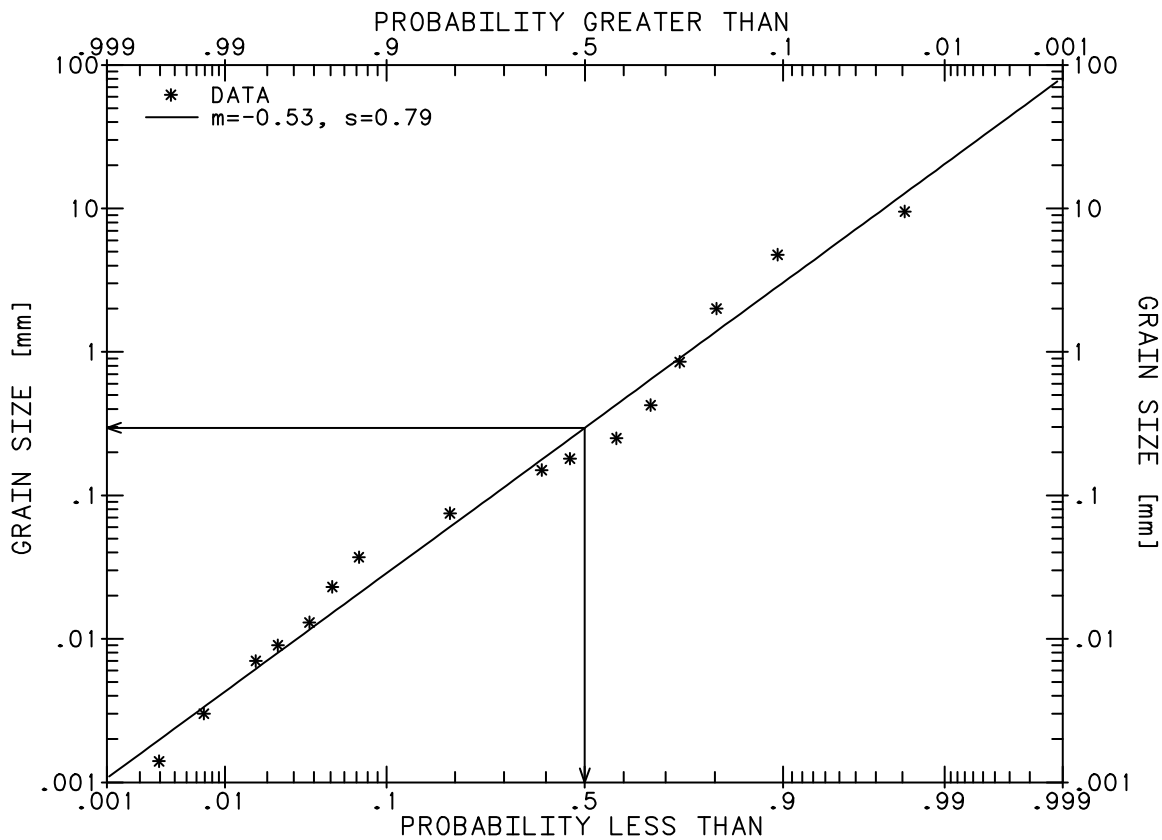


Figure 3. Grain Size Distribution

To account for a distribution of grain sizes, Wu and Gschwend's modified solution for a single grain may be multiplied by the probability distribution and integrated over the range of grain sizes. For a lognormal distribution, this becomes:

$$C(x,t) = \int_{-\infty}^{\infty} C(r,x,t) \frac{e^{-\frac{1}{2}\left(\frac{r-\mu}{\sigma}\right)^2}}{\sigma\sqrt{2\pi}} dr \quad (8)$$

While there is no closed-form solution to Equation 8, it is easily solved using Gauss quadrature. Transformation of the improper integral (infinite limits) to a proper one, necessary weights and abscissas, plus implementation are described in reference 4. The source code, input, and output may be found there as well.

7. Assumptions and Particulars of the Model

Assumptions and particulars in formulating this modeling approach for this site are as follows:

1. The medium is composed of randomly sized spheres whose diameter can be characterized by statistical measures (i.e., mean $\mu=0.30$ mm and standard deviation $\sigma=0.79$).
2. The properties of the contaminated sediments and cap (both silt) are identical and uniform ($\rho=1.5$ gm/cm³, $\varepsilon=0.5$, $\log_{10}(K)=6.6$ l/kg).
3. The source is initially contaminated uniformly ($C_0=54.8$ ppm).
4. The contaminant source is finite, having a thickness of $L=1.83$ m. The cap (isolation layer) has a thickness of 0.3048 m.
5. The solubility of the contaminant in water ($C_s=0.003$ ppm) is considered; but the solubility of the contaminant in the host (FOC) and the host in water is ignored (or beyond the original contamination level of the source). Transport of the contaminant and contaminated host in the non-aqueous phase is lumped into transport in the dissolved phase. This assumption ignores the solubility of the contaminant in the host and the host in water, but considers the solubility of the contaminant in water. This reduces the five mechanisms listed previously to four.
6. The diffusion coefficient for the contaminant (31 cm²/yr) is applied to transport of the contaminant in the aqueous phase; and the diffusion coefficient for the host (160 cm²/yr) is applied to the transport of the contaminant in the dissolved phase. Diffusion occurs within the grains and from grain-to-grain; whereas, advection occurs only from grain-to-grain.
7. The retardation factor (calculated using Equation 6) for the contaminant is applied to advection of the contaminant in the aqueous phase; and the retardation factor for the host is applied to the advection of the contaminant in the dissolved phase. No retardation factor is applied to diffusion.
8. Once the contaminant is dissolved in the host, it does not return to the aqueous phase. This assumption eliminates the necessity of formulating a relationship for the interchange of the contaminant between the aqueous and dissolved phases.

8. Model Results

The original (amorphous media/aqueous phase) model was repaired by replacing the truncated error function with the approximation plus the retardation factor calculated using Equation 6. The granular model was implemented with these properties and initial conditions, using numerical integration of Equation 8 over the grain-size distribution. Both results are shown in Figure 4.

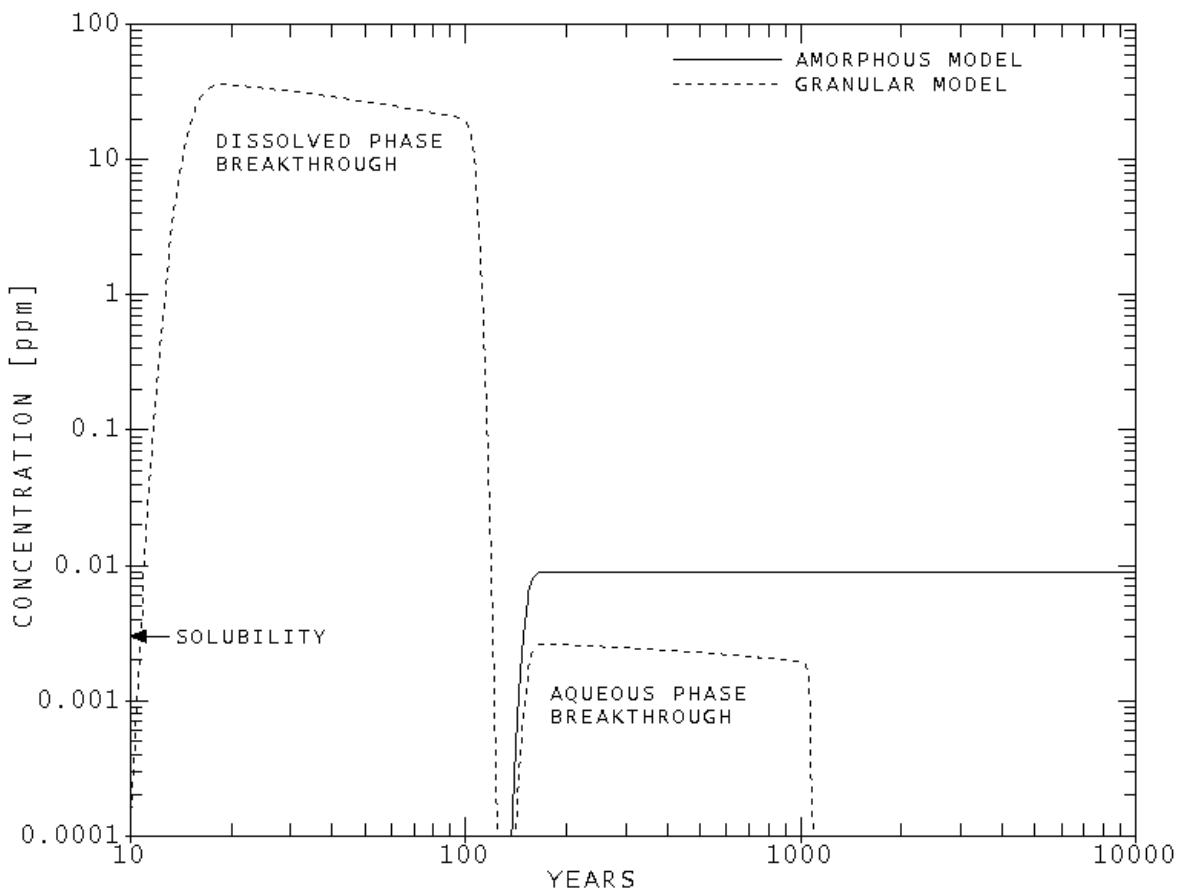


Figure 4. Model Results

The repaired amorphous model predicts a single breakthrough (the solid curve begins at about 140 years). The concentration predicted by the amorphous model is above the solubility (indicated on the left axis), in spite of its aqueous phase assumption. The granular model predicts two distinct breakthroughs, the first (dissolved phase) occurs at about 10 years and the second (aqueous phase) near 120 years). Concentration in the dissolved phase for the granular model (dashed curve) is above the solubility in water, but not in the host phase (a much lighter solvent, which facilitated deposition of the contaminant in the first place). The aqueous phase breakthrough predicted by the granular model never reaches saturation, as much of the original material has escaped by this time. Increasing the initial amount of contaminant would push the dashed curve upward toward the solubility, but never past it. Both concentrations are consistent with the model assumptions.

9. Conclusions

Even without mathematical and property errors, the original (amorphous media/aqueous phase) model used to design the barrier would have been inadequate in this case (i.e., off by more than an order of magnitude). The barrier material was not amorphous, but granular, supporting the

presence of pore water. Transport of the contaminant was not limited to the aqueous phase, because a much lighter solvent was also present and available to act as a host, facilitating rapid grain-to-grain transport. Failure of this barrier was inevitable. The capping material, per se, was not the problem, rather inadequate characterization of the transport processes led to escape of the contaminant.

Dependence on a flawed transport model resulted in an inadequate containment design. Failure of that design allowed the contaminant (a carcinogen) to escape into the environment, potentially endangering wildlife and the public. Subsequent remediation was exacerbated by failure of the original cap. Fixing the flawed design was protracted and far more costly than an effective initial design would have been. This debacle could have been avoided with accurate computational simulation of pollution dispersion.

Disclaimer: The chemical species, actual breakthrough time, location, parties, dates, and regulatory agency have been omitted from this presentation. While these details are each significant in their own right, they are not essential for this discussion of contaminant transport modeling.

Note: This author did not select any of the numerical values (i.e., properties, dimensions, durations, or concentrations) used in this analysis. These parameters were all determined by competent and experienced geohydrologists before my involvement with the project. After "fixing" the mathematics, the results were substantially in agreement with the observations, so there was no reason to question these parameters. My contribution to this effort was entirely mathematical.

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