MODELING TRANSPORT OF PCBS IN A SORBING AND RETARDING MEDIUM

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ABSTRACT

A model is developed for the transport of PCBs in a sorbing and retarding finite porous medium. The model is transient and one-dimensional. Both diffusion and simple advection are modeled. The model utilizes statistical measures of grain size in the medium.

NOMENCLATURE

- *C* concentration [moles/m³]
- C_0 initial concentration [moles/m³]
- C_a aqueous (porewater) concentration [moles/m³]
- C_m concentration in porous medium [moles/m³]
- C_X maximum concentration over time [moles/m³]
- D diffusion coefficient [m²/sec]
- D_a diffusion coefficient for the contaminant in water
- D_m diffusion coefficient for the contaminant in the porous medium
- *F* probability distribution function [dimensionless]
- G grain size G = log(2R) [log(mm)]
- G_M mean grain size [log(mm)]
- *H* hydraulic head [m]
- J flux [moles/m²/sec]
- k_H hydraulic conductivity [m/sec]
- k_T thermal conductivity [W/m/°C]
- *L* length of contaminated zone [m]
- L_i thickness of the isolation layer
- *P* cumulative probability [dimensionless]
- Q_H hydraulic (fluid) flux [m³/m²/sec]
- Q_T thermal (heat) flux [W/m²]
- *r* radial distance from center [m]
- *R* distance along *x* to reflection or radius [m]
- *T* temperature [°C]
- t time [sec]
- t_X time at which maximum concentration occurs [sec]
- *V* advective velocity [m/sec]
- x spatial location [m]

<u>Greek</u>

- ε porosity
- **R** retardation factor
- σ standard deviation of grain size [log(mm)]

INTRODUCTION

Modeling of the transport of contaminants in groundwater has many applications. Groundwater systems are often quite complex; and many sophisticated numerical models are available. A sophisticated numerical model is not always practical or necessary. A simplified approach may provide sufficient information for management strategies or provide a basis for the selection of a sophisticated numerical model. A simplified approach based on transient onedimensional diffusion in a finite medium is expanded to incorporate statistical measures of grain size in the medium and simple advection.

MODEL DEVELOPMENT

The model is developed beginning with the most simple concept of diffusion in an infinite Cartesian coordinate system and then transformed into a semi-finite Cartesian coordinate system. The length of the finite sub-domain is found to be critical. The model is then transformed from Cartesian coordinates to spherical in order to model granular media. The distribution of grain size is also included. The transport is separated into two distinct processes: diffusion and advection. The intra-grain sorption process is assumed to be diffusion-dominated; whereas, the inter-grain transport is assumed to be advection-dominated. In this model diffusion limits how much of the contaminant is available for advection. Contaminated sediment is modeled as a distributed source. The model is first developed without consideration of saturation or competition for receptor sites, which would be a worst-case scenario; as both of these effects would tend to diminish down-gradient concentration. Saturation and competition for receptor sites will be added subsequently.

<u>1D Transient Diffusion in an Infinite Medium</u></u>

The simplest model for contaminant transport would be one-dimensional transient diffusion in an infinite medium having uniform properties. The governing partial differential equation for mass transfer is given by Equation 1

$$\frac{\partial C}{\partial t} = \frac{\partial}{\partial x} \left(D \frac{\partial C}{\partial x} \right) \tag{1}$$

where *C* is the concentration, *D* is the diffusion coefficient, *t* is time, and *x* is the spatial location. For the purposes of this development, the medium will be divided at x=0. At time, t=0, the medium for x<0 is initially contaminated with a uniform concentration of $C=C_0$, and for x>0 with a uniform concentration of C=0. The medium extends infinitely far along *x*. The solution to this problem is given by Equation 2.

$$C(x,t) = \frac{C_0}{2} erfc\left(\frac{x}{2\sqrt{Dt}}\right)$$
(2)

Here erfc() is the complimentary error function. This solution is shown in Figure 1 for a range of times and locations for a diffusion coefficient, D=1.

<u>1D Transient Diffusion in a Semi-Finite Medium</u></u>

It is clear from Equation 2 and Figure 1 that the ultimate profile at infinite time is a uniform concentration of 0.5. The initially contaminated medium for all x < 0 constitutes an infinite source; whereas, the initially uncontaminated medium for all x > 0 constitutes an infinite sink. Practical applications must consider a finite source. As the one-dimensional transient diffusion equation for an infinite medium having uniform properties is linear, the solution for a finite medium can be constructed using superposition. The actual medium is approximately semifinite, that is, the initially contaminated zone is finite and the initially uncontaminated zone is infinite. In this case the applicable superposition is a reflection at $x=\pm R$. This makes the initial concentration, C=0 for x < -R, $C=C_0$ for -R < x < R, and C=0 for x > R. The superimposed solution is given by Equation 3. Concentration profiles at various times predicted by Equation 3 are shown in Figure 2, where $C_0=1$ and R=3.

$$C(x,t) = \frac{C_0}{2} \left[erfc\left(\frac{x-R}{2\sqrt{Dt}}\right) - erfc\left(\frac{x+R}{2\sqrt{Dt}}\right) \right]$$
(3)

Figure 2 shows the peak concentration, which is always at x=0, diminishing with time and the concentration profile flattening out as the finite initial contamination diffuses out into the infinite medium. Another result of this modification is illustrated in Figure 3. Figure 3 shows the concentration at a single point, x=2, over time. The concentration predicted for an infinite source (Equation 2) is shown by the dotted curve. The concentration predicted for a finite source (Equation 3) is shown by the solid curve. The infinite source solution does not exhibit a maximum; instead, it asymptotically approaches C=0.5. The finite source solution exhibits a clear maximum. The timing and magnitude of the maximum is of particular interest when considering contaminant transport. Both are dependent on the distance to the reflection, R, or the effective length scale of the contaminated medium.

The time corresponding to the maximum concentration can be found by differentiating Equation 3 with respect to time and solving for the root, as in Equation 4. The root of Equation 4 is given by Equation 5. The maximum concentration over time is then found by using this value in Equation 3.

$$xe\left(\frac{-x^2}{4Dt}\right) = (x+R)e\left[\frac{-(x+R)^2}{4Dt}\right]$$
(4)

$$t_{X} = \frac{R(2x+R)}{4D\ln\left(\frac{x+R}{x}\right)}$$
(5)

The maximum concentration and corresponding time are shown in Figures 4 and 5, respectively, for a range of Rs. These figures show that the reflection distance, R, or the effective length scale of the contaminated medium has considerable impact on the maximum concentration and corresponding time. This is why accurate characterization of the medium is critical in developing contamination management strategies.

Transformation to Spherical Coordinates

Treating the initially contaminated medium as a monolithic slab results in the largest maximum concentration and the longest time to reach that maximum concentration. A more realistic model for the initially contaminated medium is one composed of variably-sized spheres whose distribution of diameters can be described statistically. The governing partial differential equation for transient diffusion in spherical coordinates is given by Equation 6.

$$\frac{\partial C}{\partial t} = \frac{1}{r^2 \partial r} \left[r^2 D \frac{\partial C}{\partial r} \right]$$
(6)

Here r is the radial distance from the center of the grain. The solution to Equation 6 can be expressed by an infinite series, Equation 7.

$$C(r,t) = serfc\left(\frac{r}{2\sqrt{Dt}}\right)$$

$$serfc(s) = 1 - s + \frac{s^3}{6} - \frac{s^5}{30} + \frac{s^7}{168} - \frac{s^9}{1080} + \frac{s^{11}}{7920} - \frac{s^{13}}{65520} + \frac{s^{15}}{604800} - \frac{s^{17}}{6168960} + \dots$$
(7)

The infinite series in Equation 7 might be referred to as the complementary spherical error function. Superposition can then be used to create the correct boundary equations, arriving at Equation 8. Figure 6 shows the same calculations as Figure 2, except corrected for spherical spreading.

$$C(r,t) = \frac{C_0}{2} \left[serfc\left(\frac{r-R}{2\sqrt{Dt}}\right) - serfc\left(\frac{r+R}{2\sqrt{Dt}}\right) \right]$$
(8)

Figure 7 shows the concentration over time at two locations each for the Cartesian and spherical equations, respectively. The dotted curve is the Cartesian solution at X=0; and the solid curve is the spherical solution at R=0. These two curves begin at the same concentration, C_0 ; but the concentration for the spherical solution drops off much more rapidly due to spreading. The dashed curve is the Cartesian solution at X=2; and the chain curve is the spherical solution at R=2. These curves also begin at the same point, with the spherical curve dropping off sharply. In both cases the curves at the two locations asymptotically merge; because this small separation distance becomes trivial as the diffusive front advances; and the respective concentration profiles flatten. The trends shown in Figure 7 are consistent with Treybal's Figure 4.2 curves for a slab and a sphere, reproduced here as Figure 8. The horizontal axis in Figure 8 is equivalent to Dt/R^2 , or the inverse square of the term inside the error function. The vertical axis in Figure 8 is normalized concentration. The curves appear similar when Figure 8 is rotated 90° clockwise and flipped over.

Distributed Grain Sizes

Figures 4 and 5 show that the maximum concentration and the time to reach that maximum depends on the grain radius. The model for the sorption process accounts for the

variation with grain size through Equation 8. It is assumed that the distribution of grain sizes can be approximated through statistical means. Figure 9 shows grain diameter vs. probability for 8 samples taken from the same area. Significant variability is evident even between samples taken in close proximity. This variability contributes to the uncertainty of the results.

A normal probability distribution produces a straight line on a probability axis scale as constructed in Figure 9. The value where the curve intersects a probability of 0.5 is the mean. The slope of the line is always positive and proportional to the standard deviation. The normal probability distribution function is given by Equation 9.

$$F(G) = \frac{e^{-\frac{1}{2} \left(\frac{G - G_M}{\sigma}\right)^2}}{\sigma \sqrt{2\pi}}$$
(9)

Where G_M is the mean grain size, σ is the standard deviation, and F(G) is the probability distribution of grain size G. The cumulative probability, or the axis in Figure 9 labeled *Probability Less Than*, for some grain size, G, is the integral of the probability distribution from $-\infty$ to G, or Equation 10, which can be resolved to the error function.

$$P(G) = \int_{-infinity}^{G} \frac{e^{-\frac{1}{2}\left(\frac{G-G_m}{\sigma}\right)^2}}{\sigma\sqrt{2\pi}} dG = \frac{1}{2} \left[1 + erf\left(\frac{G-G_M}{\sigma\sqrt{2}}\right) \right]$$
(10)

Where P(G) is the cumulative probability of grain size *G*. A best-fit normal probability distribution is obtained for Sample 8 with a mean grain size of 0.30 mm (note: $0.30=10^{-0.53}$) and a log standard deviation of 0.79, as shown in Figure 10.

Distributed Concentration

Equation 6 with uniform properties is linear; therefore, a statistical composite concentration involving many grains can be constructed by superposition using Equation 8. The result is given by Equation 11, which must be integrated numerically.

$$C(r,t) = \frac{C_0}{2} \int_{-infinity}^{infinity} \frac{e^{-\frac{1}{2} \left(\frac{G-G_M}{\sigma}\right)^2}}{\sigma \sqrt{2\pi}} \left[serfc\left(\frac{r-R}{2\sqrt{Dt}}\right) - serfc\left(\frac{r+R}{2\sqrt{Dt}}\right) \right] dG$$
(11)

Equation 11 forms a basis for analyzing practical problems of diffusion in granular porous media. The Sample 8 grain size data are used as an example (cf. Figure 9). The mean grain size is 0.30 mm, making the mean radius 0.015 cm. A typical diffusion coefficient of 0.00001 cm/sec is used. Figure 11 is obtained using a grain size standard deviation of 0.079. The profiles in Figure 11 are quite similar to those in Figure 2, which is the slab results. This is an important check on the asymptotic behavior of Equation 12. If the grain size is relatively large and the standard deviation is relatively small (i.e., nearly uniform grains), compared to the diffusion coefficient and characteristic time, Equation 11 should approach the slab result, or Equation 3, which it does.

If the grain size standard deviation is increased to 0.79, a significant change in the shape of the profiles can be seen, as illustrated in Figure 12. Increasing the grain size standard deviation increases the fraction of smaller and larger diameter grains relative to the mean. The contribution of the smaller grains to the integrated concentration is a more rapid decrease at the center; as the relative distance from the source is much larger. The contribution of the larger grains is to hold the concentration up farther away from the center; as the part of the concentration profile plotted is still deep within the large grains. Mass is conserved, thus, the area under each curve when weighted by the radius squared is the same. As more smaller grains *pull* the profiles down toward the center and more larger grains *stretch* the profiles out, a different shape is obtained (Figure 12) as the radial effects dominate.

Simple Advection

Advection is the transfer of mass due to the presence of a flow through the medium, in this case, porewater. Up to this point there has been no distinction between transport through the medium proper and transport through the flowing porewater. These will be distinguished by the subscripts m and a for *medium* and *aqueous*, respectively. Equation 1 can be modified to account for simple one-dimensional advection, resulting in Equation 11.

$$\frac{\partial C_a}{\partial t} = D_a \frac{\partial^2 C_a}{\partial x^2} - \frac{V \partial C_a}{\Re \partial x}$$
(12)

where V is the advective velocity and \Re is the retardation factor. The complete solution to Equation 12, which can be found using Laplace transforms, is given by Equation 13.

$$C_{a} = \frac{C_{0}}{2} \left[erfc \left(\frac{X - \frac{V}{\Re}t}{2\sqrt{D_{a}t}} \right) + exp \left(\frac{\frac{V}{\Re}x}{D_{a}} \right) erfc \left(\frac{X + \frac{V}{\Re}t}{2\sqrt{D_{a}t}} \right) \right]$$
(13)

For typical property and dimensional values, the second term in Equation 13 is negligible. Neglecting this term results in a simplified solution for advection that is a slight modification to the solution for pure diffusion, Equation 2, of adding the term Vt/\Re . This same modification can be made to Equations 3, 9, and 11.

Diffusion vs. Retardation

Diffusion is the transfer of mass due to the presence of a concentration gradient. Mathematical analysis of this important mass transfer process is based on Fick's Law of Diffusion, Equation 14. Equation 14 relates the flux, J, the gradient of the concentration, C, and the diffusion coefficient or diffusivity, D. The units of flux are moles per area per time (e.g., [moles/m²/sec]). The units of concentration gradient are moles per volume per length (e.g., [moles/m³/m]). The units of diffusivity are length-squared per time (e.g., [m²/sec]).

$$I = -D \frac{\partial C}{\partial x} \tag{14}$$

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It is important to note that Equation 14, or Fick's Law, *defines* diffusivity, in exactly the same manner as Fourier's Law, Equation 15, *defines* thermal conductivity and Darcy's Law, Equation 16, *defines* hydraulic conductivity. Each of these coefficients are experimentally determined by measuring the molar flux and concentration gradient, heat flux and temperature gradient, and fluid flux and hydraulic gradient, respectively.

$$Q_{T} = -k_{T} \frac{\partial T}{\partial x}$$
(15)

$$Q_{H} = -k_{H} \frac{\partial H}{\partial x}$$
(16)

In order to determine thermal conductivity, k_T [W/m/°C], one measures the heat flux, Q_T [W/m²] and the gradient of the temperature, $\partial T/\partial x$ [°C/m]. The ratio of heat flux to temperature gradient is the thermal conductivity by definition. In order to determine the hydraulic conductivity, k_H [m/sec], one measures the fluid flux, Q_H [m³/m²/sec] and the hydraulic gradient, $\partial H/\partial x$ [m/m]. The ratio of fluid flux to hydraulic gradient is the hydraulic conductivity by definition. In order to determine the coefficient of diffusion, D [m²/sec], one measures the molar flux, J [moles/m²/sec] and concentration gradient, $\partial C/\partial x$ [moles/m³/m]. The ratio of molar flux to concentration gradient is the diffusivity, D [m²/sec], by definition.

Equation 14 can be combined with the principle of the conservation of chemical species to form an expression for the time rate of change of the concentration, or the partial differential equation for transient molecular diffusion, Equation 1. Equation 14 *defines* the diffusivity; therefore, Equation 1 can also be used to experimentally determine the diffusion coefficient in a system where there is no advection. One need only measure the rate of change of the concentration, $\partial C/\partial t$ [moles/m³/sec], and the gradient of the concentration gradient, $\partial^2 C/\partial x^2$ [moles/m³/m²]. The ratio of these two quantities is the diffusivity, *D* [m²/sec], *by definition*.

Retardation is an advective phenomenon. The retardation factor is an empirical correction to account for an apparent time lag in the advancement of a specific substance through a specific medium. The physical phenomenon thought to be responsible for retardation is a temporary attachment of the advecting contaminant to the medium at the molecular scale. This explanation for retardation requires the presence of receptor sites in the medium that accommodate the contaminant. If there is no porous medium, then there can be no retardation. According to this conceptual model for retardation the contaminant molecules are removed from the porewater and attach themselves to receptor sites in the medium, leaving less contaminant in the porewater as it flows through the medium. When the receptor sites are filled, the addition of one contaminant molecule forces another back into the porewater, resulting in an apparent time lag.

Retardation results from the attachment and detachment of individual contaminant molecules between the porewater and receptor sites in the porous medium. Retardation does not occur within the porewater or the porous media, per se, as is the case with diffusion. Retardation and diffusion are two separate processes operating simultaneously. Retardation is an advective phenomenon. *Advection is subject to retardation; whereas diffusion is not*.

Problematic Lack of True Properties

Several problems arise in the application of these principles and equations to real-world transport of contaminants, which exhibit large retardation factors or very slow migration rates. This is the lack of true properties. A carefully controlled laboratory experiment is necessary to measure true diffusive properties. There must be no advection present in order to measure the diffusion coefficient. Constructing the experimental apparatus is further complicated by the necessity of having an undisturbed layer of the target porous media through which to measure the advance of the target contaminant. The diffusion of PCBs through silt is very slow, which means the experimental conditions must be maintained over a long period of time, perhaps years, even decades. The advective transport rate and retardation factor can be measured much more easily, provided that the diffusion is negligible -- an assumption that requires accurate diffusion data in order to justify. Under such idyllic conditions the hydraulic conductivity and retardation factor could be inferred from field data by tracking the center of mass of a finite point source (i.e., a single *spill*) containing a species that is not retarded along with the target species that is retarded. Measurement of these field data may also require years of collection.

What seems to be the conventional way of obtaining properties for use in analyzing the transport of PCBs in groundwater, is to estimate these from properties of totally different substances in solvents different from water and applying rules of thumb. This practice is questionable and introduces the possibility of errors, not in mere percent, but in orders of magnitude. If these rules of thumb are based on actual data, then why not use the real data? If the rules of thumb are not based on actual data, then there is no basis on which to validate the properties obtained by using them. This situation is further complicated in that retardation may have been incorrectly assumed in the process of generating the rules of thumb. In order to be consistent when using a diffusion coefficient that somehow erroneously included retardation, that same retardation factor must be rolled back into the calculations; but doing so must not be used as a justification that diffusion is subject to retardation, which is clearly an advective phenomenon.

Retardation and Conservation of Mass

The rate of increase of the contaminant in the porous medium is equal to the flux of contaminant into the medium less the flux leaving the medium. The governing partial differential equation for the aqueous concentration, C_a , is given by Equation 12. Conservation of mass requires that the partial differential equation for the corresponding process in the porous medium, C_m , in the presence of retarded advection be given by Equation 17:

$$\frac{\partial C_m}{\partial t} = D_m \frac{\partial^2 C_m}{\partial x^2} - \frac{V}{(1-\varepsilon)} \left(1 - \frac{1}{\Re}\right) \frac{\partial C_a}{\partial x}$$
(17)

where ε is the porosity. As the retardation factor, \Re , approaches 1, there is no retardation and no accumulation of the contaminant in the porous medium. As the retardation factor approaches infinity, all of the contaminant leaves the porewater and fills the receptor sites in the porous medium. As the porous medium. As the porous medium. As

the porosity approaches 1, the porous medium occupies none of the total volume; thus, any presence of a contaminant would correspond to an infinite concentration. This uptake of contaminant from the porewater into the porous medium by virtue of retardation is often neglected. Equation 17, by the conservation of mass, requires that the concentration of the contaminant in the porous medium must rise proportional to the retardation factor and inversely proportional to the porosity. An entering porewater concentration of 0.009 ppm, a retardation factor of 30,000, and a porosity of 0.3 could produce a concentration in the porous medium of over 900 ppm. If there were indeed sufficient receptor sites in the porous medium to produce such a retardation effect, the eventual concentration in the porous may exceed the original contaminant level. Retardation depends on available receptor sites; therefore, the retardation factor must vary with concentration. A value that might be appropriate at low concentrations may be inappropriate at high concentrations. Whether the contaminant could subsequently leave the receptor sites in the porous medium and re-enter the porewater would depend on the species and conditions. If in addition to the contaminant entering via the porewater, contaminant is initially present in the porous medium, the diffusion associated with contaminant source would add to the concentration within the porous medium resulting from retardation. As all of the simplified governing equations have been linear, this effect can be superimposed to obtain the total.

APPLICATION

An example is given to illustrate the magnitude of the quantities involved. Consider the case where the thickness of the isolation cap, L_i , is 2 feet, the advective velocity, V, is 100 ft/yr, the initial (entering) concentration in the porewater, C_0 , is 0.009 ppm, the porosity, ε , is 0.3, both diffusion coefficients, D_a and D_m , are 0.034 ft²/yr, and the retardation factor, \Re , is 100. Equation 1 can be solved analytically; however, Equation 17 has no known [yet] analytical solution; therefore a numerical solution will be used. For the numerical solution, 500 elements are used and 84,660 time steps are taken. Figure 13 shows the concentration in the porewater over time at the end of the isolation cap as computed analytically and numerically. This comparison is made in order to check the accuracy of the numerical solution. Figure 14 shows the concentration in the porewater throughout the cap at 1.46 years. Again the analytical and numerical solutions are shown for comparison. The dramatic impact of retardation on the concentration of the contaminant accumulating at the receptor sites within the porous media is shown in Figures 15 and 16. Figure 15 shows the concentration in the porewater (the dotted curve) and porous medium (solid curve) over time at the end of the isolation cap. Note that the concentration scale for Figures 15 and 16 are much greater than for Figures 13 and 14. Figure 16 shows the concentration throughout the cap at 1.46 years.

CONCLUSIONS

The figures in the application do not include any non-aqueous phase diffusion through the cap due to initial contamination of the porous medium underlying the cap. These calculations were made with a retardation factor of 100. The impact would be 300 times as great were the retardation factor 30,000. These results should not be surprising. The whole purpose of extraction systems is to remove the contaminant from the water and accumulate a high concentration in the extraction medium. When most of the receptor sites are filled, the medium is replaced and the

contaminated medium disposed of. If an in-situ isolation cap is used to prevent a contaminant from escaping and retardation is the operative mechanism, by the time the retarded front reaches the end of the cap it could be contaminated to a level requiring it to be removed and treated as hazardous waste.

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Figure 1. Concentration Profiles for Infinite Medium



Figure 2. Cartesian Concentration Profiles for Semi-Finite Medium



Figure 3. Concentration at a Point over Time



Figure 4. Maximum Concentration vs. Reflection Distance



Figure 5. Time of Maximum Concentration vs. Reflection Distance



Figure 6. Spherical Concentration Profiles at Various Times



Figure 7. Cartesian and Spherical Concentration at Two Points over Time.



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Figure 9. Measured Grain Size Probability for 8 Samples



Figure 10. Best Fit Normal Probability Distribution for Sample 8



Figure 11. Statistically Averaged Spherical Concentration Profiles for $\sigma=0.079$



Figure 12. Statistically Averaged Spherical Concentration Profiles for $\sigma=0.79$



Figure 13. Porewater Concentration History at X = 2 Feet



Figure 14. Porewater Profile at T = 1.46 Years



Figure 15. Porous Medium History at X = 2 Feet



Figure 16. Porous Medium Profile at T = 1.46 Years