Predicted Retardations of Concentration Fronts Using a Mass-Balance Approach

by Ronald K. Stoessell^a

Abstract

Regardless of the chemical controls for a pollutant on sediment sorption or release from an immiscible organic liquid, a simple mass balance can be used to predict the retardation of the ground water pollutant front moving downstream from a source of constant composition. The retardation is the ratio of the total molar difference of pollutant across the front to the molar difference in ground water. The mass-balance approach presented here uses a flushing factor for the entire front. It is implicit in the commonly used retardation factor for pollutants undergoing linear sorption, but does not appear to have been used to predict retardation of concentration fronts involving nonlinear sorption or partitioning in hydrochemical studies. Yet the computations require only limited experimental or field data or the use of an algorithm describing sorption or partitioning of the pollutant. Close agreement was obtained between retardations predicted using the mass-balance approach and those reported from numerical modeling and column experiments in the literature.

Introduction

The downstream movement of a pollutant from a source results in a concentration front as shown in modeling and laboratory experiments (Newman et al. 1991; Appelo 1994; Fetter 1993; Domenico and Schwartz 1997; Drever 1997) and in the use of chromatography for laboratory analyses (Appelo 1996). Field examples, unrelated to pollution, include redox-controlled uranium roll-fronts in sandstones (Campbell 1977) and salinity fronts developing from ground water movement in coastal aquifers (Appelo 1996).

This paper describes a simple procedure for estimating the maximum mobility of a pollutant undergoing sorption, by predicting the retardation of a concentration front moving away from a source of constant composition. The procedure follows from mass constraints that are summarized by Appelo (1996), "the front position can be calculated from the integral mass balance." The reader is referred to this review paper for a literature review of chromatography as applied to natural systems.

The procedure is illustrated in this paper by comparing the retardation of fronts determined from modeling and column experiments with those predicted using the mass-balance approach. Although the mass-balance procedure presented here is implicit in the commonly used retardation factor for pollutants undergoing linear adsorption, it does not appear to have been used to predict retardation of pollutants undergoing nonlinear sorption or partitioning in hydrochemical studies.

Retardation of a Concentration Front Due to Sediment Sorption

The general equation form of the retardation of a concentration front follows directly from a mass balance across the front. The shape and width of the front are not given by the retardation. These factors are controlled both by the sorption process and by dispersion and diffusion.

The concentration front moves downstream by replacing the downstream ground water with the source ground water and the downstream moles of a pollutant on sediment sites with those in equilibrium with the source fluid. If the sediment composition is homogeneous, a constant difference in pollutant concentrations exists between the area upstream of a front and that downstream of the front. If the ground water velocity is constant and the aquifer is isotropic, the rate of movement of the front will be constant.

The retardation of the position of the center of mass of the concentration front is expressed as a mass balance across the front. Per liter of sediment pore space, the front retardation, $R_{\rm f}$, for sorption is the ratio of the moles of pollutant being placed on or removed from sediment sites and the ground water to the moles in 1 L of ground water passing through the front. This ratio is computed using the concentration differences between the area upstream of the front and the area downstream of the front. For a source of constant composition, $R_{\rm f}$ of the pollutant front can be written as

$$R_{f} = [(M_{B,source} - M_{B,downstream}) + (q_{B,source} - q_{B,downstream})]$$

$$/(M_{B,source} - M_{B,downstream})$$
(1)

where $q_{B,source}$ and $q_{B,background}$ are the moles of B per liter of pore volume in the sediment in equilibrium with source and back-

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ground molarities, respectively, $M_{B,source}$ and $M_{B,background}$. In the absence of multiple fronts, $q_{B,downstream}$ always equals $q_{B,background}$; however, as discussed later, $M_{B,downstream}$ is not always equal to $M_{B,background}$.

A convenient form of Equation 1 is

$$R_{f} = 1 + (q_{B,source} - q_{B,downstream})/(M_{B,source} - M_{B,downstream})$$
 (2)

The term $(q_{B,source} - q_{B,downstream})/(M_{B,source} - M_{B,downstream})$ is the flushing factor of Appelo and Postma (1993; Equation 11.4) written for the entire front. Those authors derive Equation 2 from the advection and sorption partial differential equation (with dispersion set to zero) in which the flushing factor is written in partial differential form as dq_B/dM_B . They then substitute the derivative of the appropriate isotherm describing sorption for the flushing factor. However, dq_B/dM_B at any point along the front will not necessarily equal the gradient across the entire front, and the latter is used in Equation 2 to calculate the retardation of the front.

For a charged pollutant B^b undergoing sediment sorption, Equation 2 becomes

$$R_{\rm f} = 1 + [L\rho_{\rm sed}/(\theta \mid b \mid)](X_{\rm B,source} - X_{\rm B,downstream})/(M_{\rm B,source} - M_{\rm B,downstream})$$
(3)

where $X_{\rm B}$ is the equivalent-site fractions of B^{b+} on sediment sites. The factor ${\rm L}\rho_{\rm sed}/(\theta\,|\,b\,|)$ converts $X_{\rm B}$ to moles of B per liter of pore space. L, $\rho_{\rm sed}$, θ , and $|\,b\,|$, respectively, are the equivalents of sites available per kilogram of sediment, the density of dry sediment in kg/L, the sediment porosity fraction, and the absolute charge of pollutant B.

For an uncharged pollutant B undergoing sediment sorption, Equation 2 becomes

$$R_{\rm f} = 1 + [Q\rho_{\rm sed}/\theta](X_{\rm B,source} - X_{\rm B,downstream})/(M_{\rm B,source} - M_{\rm B,downstream})$$
(4)

where X refers to the mole fraction of sites and Q is the total number of moles of available sites per kilogram of sediment.

The reciprocal of R_f multiplied by the ground water velocity gives the downstream velocity of the front. This velocity is constant in a homogeneous, isotropic aquifer with a constant concentration source and is a maximum estimate of the mobility of a pollutant undergoing adsorption. Precipitation, decay, and a decrease in the source concentration of a pollutant will slow the downstream pollutant movement.

The concentrations needed in Equations 3 and 4 can be determined by experiments or measured in the field. $X_{\rm B, source}$ or $X_{\rm B, source}$ is that determined from a batch experiment equilibrating an excess amount of source ground water with a trace amount of sediment. $X_{\rm B, downstream}$ or $X_{\rm B, downstream}$ can be determined in a similar experiment using background ground water. $M_{\rm B, downstream}$ can often be estimated if it differs from $M_{\rm B, background}$. If field data are available, these concentrations can be determined by direct measurement on sediment upstream of the front and on downstream sediment and ground water. Alternatively, if the sorption model is known for the pollutant, the model can predict pollutant sediment concentrations in equilibrium with source ground water and downstream ground water.

If $M_{B,background}$ is zero, $M_{B,downstream}$ is also zero in the absence of multiple fronts. If B is competing with other components for exchange sites and the background B concentration on the sediment is non-zero, $M_{B,downstream}$ may differ from $M_{B,background}$ due to trace

amounts of exchange moving ahead of the front. However, trace concentrations are not expected to make a significant difference in the predicted retardation from Equations 3 and 4.

A major change in molarity between the source and background ground water of a component competing with B for sorption sites with the pollutant, combined with non-zero background B concentrations on sediment, should result in a significant difference between M_{B.downstream} and M_{B.background}. The downstream pollutant and site-competing component adjust their molarities (through site exchange) to reestablish the equilibrium ratio with their background concentrations on the sediment sites. If the pollutant had only minor site occupancy on the sediment, the downstream molarity of the competing component approaches that of its source molarity and M_{B.downstream} can be calculated from a sorption model. If the pollutant had major site occupancy on the sediment, $M_{B,downstream}$ can be obtained by modeling the passage of the initial fluid pore volume moving downstream away from the source until M_B becomes constant. Alternatively, M_{B,downstream} can be determined from field measurement downstream of the plume.

Retardation of a Concentration Front Due to Partitioning Between an Immiscible Organic Fluid and Ground Water

The same type of mass balance used for sorption in Equation 2 can also be written for the release of an organic pollutant B into ground water from a dispersed immiscible organic fluid in the sediment, e.g., toluene in gasoline being partitioned with ground water. For a source of constant composition,

$$R_f = 1 + r(OF_{B,source} - OF_{B,downstream})/(M_{B,source} - M_{B,downstream})$$
 (5

where r is the volume ratio of organic fluid to ground water within the pore space and ${\rm OF_B}$ is the moles of B per liter of organic fluid. ${\rm OF_{B,downstream}}$ and ${\rm M_{B,downstream}}$ can be directly measured from samples of ground water and the organic fluid in the aquifer, prior to flushing. If ${\rm M_{B,source}}$ is zero then ${\rm OF_{B,source}}$ is zero at equilibrium. ${\rm OF_{B,upstream}}$ and ${\rm M_{B,upstream}}$ can replace ${\rm OF_{B,source}}$ and ${\rm M_{B,source}}$ in Equation 5 if a residual amount of B in the organic fluid persists due to a lack of equilibrium. This was the case in the example used in this study. If the flushing fluid contains some of B, ${\rm OF_{B,upstream}}$ can be measured in a partitioning experiment of an excess amount of the flushing fluid with a trace amount of the organic fluid. Alternatively, if the partition model is known for the pollutant, the model can predict B concentrations in the organic fluid in equilibrium with source ground water and downstream ground water.

Isotherms

In this paper, retardations determined from modeling and column studies are compared with those predicted from the mass-balance approach procedure. The sorption isotherms described in this section are those whose parameters were reported in these studies by fitting the laboratory data used in the retardation comparisons. The concentration units normally used with the Distribution, Freundlich, Langmuir, and Modified Langmuir isotherms are $S_{\rm B}$ in mg/kg of solid and $C_{\rm B}$ in mg/L of solution. In terms of these units, Equation 2 becomes

$$R_f = 1 + (S_{B,source} - S_{B,downstream}) \rho_{sed} / \theta] / (C_{B,source} - C_{B,downstream})$$
 (6)

These same isotherms are also used to describe partitioning of an organic pollutant B between an immiscible organic fluid and ground water, e.g., the commonly used partition coefficient that is equivalent to the Distribution isotherm. For partitioning, the concentration of B in the organic fluid is usually given in mg/L as $s_{\rm B}$. Equation 5 becomes

$$R_{f} = 1 + r(s_{B,source} - s_{B,downstream}) / (C_{B,source} - C_{B,downstream})$$
 (7)

The following isotherms are given in terms of S_B for sediment sorption; however, substitution of s_B for S_B produces the algorithm used to fit experimental data for partitioning between ground water and an immiscible organic liquid.

The simplest function is the linear Distribution isotherm with a single fit parameter, K_D , the Distribution coefficient.

$$S_B = K_D C_B \tag{8}$$

The retardation can be directly computed in situations where sorption follows the linear Distribution isotherm. Substitution of Equation 8 into Equation 6 yields the well-known relation,

$$R_{f} = 1 + K_{D} \rho_{sed} / \theta \tag{9}$$

The usual problem with using Equation 9 is that sorption is not a linear process, so K_D varies with solution composition.

The Freundlich isotherm contains two fit parameters, K_F and n. The parameter n usually varies between 0.7 and 1.1 (Langmuir 1997; Appelo 1999). K_D is a special case of the Freundlich isotherm when n is one.

$$S_B = K_F(C_B)^n \tag{10}$$

Typically, in the literature (Rao and Davidson 1979; Fetter 1992; Appelo and Postma 1993), dS_B/dC_B replaces ($S_{B,source}-S_{B,downstream}$)/($C_{B,source}-C_{B,downstream}$) in Equation 6. Use of Equation 10 then gives

$$R_f = 1 + (\rho_{sed}/\theta) K_F n(C_B)^{(n-1)}$$
 (11)

Because the concentration is changing in a nonlinear fashion across the concentration front, the equation will not necessarily predict $R_{\rm f}$ correctly by using the midpoint concentration for $C_{\rm B}$ of the front. The same result applies, of course, to the other nonlinear isotherms defined next, for which similar expressions can be derived.

The Langmuir isotherm contains the fit parameter, k, and the maximum amount of sorbed solute, S_{max} .

$$S_B = S_{max}kC_B/(1 + kC_B)$$
 (12)

The Modified Langmuir isotherm (Newman et al. 1991) contains S_{max} and two fit coefficients K_{ML} and β . Newman et al. called

Table 1 Literature $R_{\rm f}$ Compared to Predicted $R_{\rm f}$ for Sorption on Sediment										
Model or Experiment	R _f from Study	C^ Source or Downstream mg/1 _{aq}	S^ Equil. mg/kg _{sed}	ρ _{sed} /θ kg _{sed} /1 _{aq}	R _f Pred. Equation 6					
Newman et al. (1991)	modeling adsorption									
Li										
Freundlich Langmuir	34.1 27.0	100 100	136 103*	24.3 24.3	34.0 26.0					
Modified Langmuir	29.2	100	116	24.3	29.2					
Voudrias et al. (1993)	column adsorpt. and desorpt.									
Cs										
Mudstone Mudstone	3.4 and 2.6# 3.6 and 2.6#	1.33 1.33	1.48 1.48	1.78 1.78	3.0 3.0					
Rao and Davidson (1979) 2-4 D amine	column adsorption									
Cecil soil	1.2	5000	764	1.17	1.2					
Eustis soil	1.1	5000	381	1.17	1.1					
Bürgisser et al. (1993)	column adsorpt. and desorpt.									
Cd										
Cristobalite Methylene blue	2.8 and 2.6	1.97	1.34	2.82	2.9					
Cristobalite	3.4 and 3.8	3.17	3.02	2.82	3.7					

[^]C is either the aqueous source concentration during sorption in which the downstream concentration was zero or is the downstream concentration in desorption in which the source concentration was zero. S is the sediment concentration in equilibrium with C.

^{*}S_{Li} was taken from a plot of the Langmuir isotherm.

 $^{^{\}text{\#}}$ Column R_f was based on the center of mass of the wavefront.

this function the Modified Freundlich isotherm; however, it reduces to the Langmuir isotherm when β is unity.

$$S_B = S_{max} (K_{ML} C_B)^{\beta} / (1 + (K_{ML} C_B)^{\beta})$$
 (13)

None of the aforementioned isotherms explicitly accounts for sorption due to exchange reactions. Readers are referred to Langmuir (1997; Table 10.8) for a convenient compilation of 25°C and 1 atm binary cation-exchange data on clays in the form of a Power-Exchange function. This function can be derived from the massaction law of a binary-cation exchange reaction using the Gapon convention together with assumptions on maintenance of local electrostatic balance and the availability of exchange sites for mixing (Stoessell 1998). Other authors prefer to use the Gaines-Thomas or Vanselow conventions in describing exchange reactions involving competition for sites. A discussion of those conventions and the resulting cation-exchange equations can be found in Sposito (1981; Chapter 5) and Appelo and Postma (1993; Chapter 5).

Predicted Retardations for Sorption Compared to Those from the Literature

Retardations of concentration fronts from selected studies of sediment sorption of modeling and column experiments are compared in Table 1 with predicted retardations. Unless otherwise noted, the literature values of $R_{\rm f}$ were computed from plots of concentration fronts using the position of the mid-point concentration. Note that as a concentration front increases in asymmetry, the mass center will shift from the position of the mid-point concentration, producing some error in comparison with a predicted value from Equation 6, which is for the center of mass of the front. Unless otherwise noted, the literature values of $R_{\rm f}$ were either the ratio of the time for passing the pollutant's concentration front to that of a nonsorbed tracer or the number of pore volumes needed to pass the pollutant's concentration front through a column.

The predicted R_f values from Equation 6 in Table 1 assumed zero pollutant concentrations downstream and upstream, respectively, of the concentration fronts for adsorption and desorption. Predicted R_f values were identical for both adsorption and desorption in Table 1. For each predicted R_f , the source value C for adsorption or background value C for desorption, the corresponding equilibrium value S, and ρ_{sed}/θ for the sediment are listed in Table 1. S was either calculated from a fitted isotherm algorithm or taken directly from an isotherm plot. Equation 6 was applied to column experiments in which the source concentration was maintained constant long enough so that the outlet concentration reached the source concentration, i.e., a complete breakthrough curve is obtained.

Comparison with Numerical Modeling

Newman et al. (1991) fitted experimental sediment sorption data of lithium with the Prow Pass Tuff for Equations 8, 10, 12, and 13 (the Distribution, Freundlich, Langmuir, and Modified Langmuir isotherms, respectively). The parameters for each of these models are listed in their Table 5. They used these fits to numerically model the concentration fronts based on transport simulations using the advective-dispersive equation with sorption equilibrium. In their Figure 5, the breakthrough curves for a constant source concentration of 50 mg/L are shown as a function of time for lithium for each of the presented models as well as for a conservative tracer.

The ratio of ρ_{sed}/θ used in modeling was not given by Newman et al. (1991). For use in Equation 6, this ratio was back-calculated from Equation 9 as 24.3, using the computed R_f of 11.33 from their modeling with the linear Distribution isotherm combined with their measured K_D of 0.425 L/kg of sediment. Newman (1998) told the author that the tuff had a porosity of 10% so the bulk-sediment density would have to be 2.43 g/cm³ with a corresponding grain density of 2.7 g/cm³.

 $R_{\rm f}$ of the breakthrough curve for each model is given in Table 1 together with the predicted $R_{\rm f}$ value from Equation 6. The agreements between modeled and predicted $R_{\rm f}$ values are exact for the Freundlich and Modified Langmuir isotherms. The minor 4% disagreement between the modeled and predicted $R_{\rm f}$ for the Langmuir isotherm may be a reflection of the higher asymmetry of the concentration front (see their Figure 5). The predicted $R_{\rm f}$ for the Langmuir isotherm in Table 1 used $S_{\rm Li}$ taken from a plot of the Langmuir isotherm fit to their data (their Figure 4). Use of the $S_{\rm max}$ (their b parameter) in their Table 5 for the Langmuir isotherm predicted a $S_{\rm Li}$ of 76 mg/kg in equilibrium with 100 mg/L of lithium, inconsistent with their Figure 4.

Comparison with Column Measurements for Sediment Sorption

Voudrias et al. (1993) presented experimental column adsorption and desorption breakthrough curves for cesium in a brine-saturated mudstone. R_f values for sorption and desorption are given in their Table 7 and in Table 1 of this study. The predicted R_f values in Table 1 used C_{Cs} and ρ_{sed}/θ in their Table 2 with S_{Cs} computed using Equation 10 with the Freundlich isotherm parameters in their Table 3.

The agreements for R_f are good, 3.0 predicted versus 3.4 and 3.6 from the column experiments (see their Table 7) using the wavefront positions of 50% of the maximum cesium concentration. Use of the center of mass of the experimental breakthrough curves to compute R_f gives a value of 2.6 (see their Table 7). The three column R_f values bracket the predicted value.

Rao and Davidson (1979) presented breakthrough curves in their Figures 1, 2, and 3 for 2,4-D amine and a conservative tracer in Webster, Cecil, and Eustis soils, respectively. The measured values in Table 1 are from the 5000 mg/L breakthrough curves. Breakthrough curves were also given for source concentrations of 50 mg/L; however, those breakthrough curves were incomplete, indicating the source concentration was not maintained constant long enough to use Equation 6.

Rao and Davidson (1979) did not give ρ_{sed}/θ values for the three soils. Each soil had been air dried and packed in small increments in glass cylinders. The assumption was made that the porosity of the soils was approximately the same because of the similar packing procedure and that the density of mineral grains was also the same because most of the common minerals in soils have similar densities. ρ_{sed}/θ was back-calculated as 1.17 for the Webster soil using the measured retardation of 1.42 (from their Figure 1) in Equation 6. This ratio was then used in Equation 6 for the Cecil and Eustis soils. The equilibrium S concentrations in Table 1 were computed using the fitted Freundlich isotherm parameters (for each soil) in their Table 2.

The agreement is exact between the column and predicted retardations for sorption for the Cecil and Eustis soils in Table 1. The ratio of ρ_{scd}/θ used in the predicted retardations was lower than expected, but the results are certainly consistent with the measured retardations.

Table 2	
Literature R _f Compared to Predicted R _f for Release of a Pollutant from an Immiscible Organic	Fluid

Wise et al. (1992)		C	S Downstream mg/l _{aq}	Upstream* mg/l _{org}	Downstream mg/ l _{org}	r l _{org} /l _{aq}	R _f Pred. Equation 7
Experiments Freundlich	R _f column	Upstream* mg/l _{aq}					
benzene	41	.010	22.99	16.7	6002	0.156	42
toluene	150	.067	40.73	235	39,665	0.156	152
ethylbenzene	409	.002	2.91	9	7766	0.156	417
meta-xylene	347	.041	13.12	155	29,441	0.156	350
para-xylene	504	.041	1.03	181	3407	0.156	510
ortho-xylene	397	.003	4.49	16.1	11,592	0.156	403

^{*}The upstream concentrations were substituted for source concentrations in Equation 7 because an apparent lack of equilibrium resulted in organics persisting in the upstream ground water after passage of the front.

Bürgisser et al. (1993) show Cd and methylene blue breakthrough curves for adsorption and desorption, respectively, in their Figures 4 and 7. The two input solutions were a 17.5 μM Cd solution and a 9.91 μM methylene blue solution, which displaced a 10 mM NaNO $_3$ solution in columns packed with cristobalite sand. Two different flow velocities produced identical breakthrough curves in Figure 4, suggesting equilibrium in the ion-exchange process of Cd replacing Na. R_f values for Cd and methylene blue were computed from the breakthrough curves in their Figures 4 and 7, respectively. Predicted R_f in Table 1 used S_{Cd} and $S_{methylene blue}$ taken from batch equilibration data in the isotherms in their Figures 6 and 8, respectively.

The column R_f for Cd of 2.8 for adsorption and 2.6 for desorption are in good agreement with the predicted R_f of 2.9. The predicted R_f for methylene blue of 3.7 was bracketed by the column R_f of 3.4 for adsorption and 3.8 for desorption. In both cases the least asymmetrical breakthrough curve produced the closest column R_f to the predicted R_f .

Comparison with Column Measurements for Release of a Pollutant from an Organic Fluid

Wise et al. (1992) reported breakthrough curves from Borden and Kao (1989) for the release of organic constituents into water from gasoline that had been dispersed into sand. The column R_f values in Table 2 were computed from the mid-point position of the breakthrough curves shown in their Figures 8a (benzene), 9a (toluene), 10a (*meta*- and *para*-xylene), and 11a (*ortho*-xylene).

The column contained 4.6 mL of immobile gasoline within a total pore space of 34 mL, making r in Equation 7 equal to 0.156. Wise et al. (1992) fitted the sorption data from Borden and Kao (1989) for each of these organic components to the Freundlich isotherm (Equation 10), modified for representing partitioning as $s_B = K_F(C_B)^n$. The Freundlich parameters and the upstream and downstream aqueous concentrations were listed in their Table 1 and used to calculate s_B for each of the organic constituents. Upstream concentrations were substituted for source concentrations in Equation 7 because trace amounts of organics persisted in ground water due to a lack of equilibrium.

The predicted R_f values from Equation 7 are shown in Table 2. Agreements between column R_f and predicted R_f are each within 1%.

Summary

The predicted retardations of the concentration fronts in Table 1 and Table 2 show good to excellent agreement with those determined by numerical modeling and from column experiments for conditions of constant source concentrations. The use of Equation 6 for sediment sorption and Equation 7 for partitioning with an organic liquid does not require specific knowledge of the chemical controls over either processes.

The only required chemical data for Equations 6 and 7 are the equilibrium pollutant upstream and downstream concentrations. These data can be determined from field measurements and/or simple equilibration experiments. If the chemical controls on sorption and partitioning are known, the chemical data can be calculated from functions describing those chemical controls.

The author wants to emphasize that this method is based on conservation of mass and is an application of the work of many authors, e.g., the integral mass-balance approach of Appelo (1996) and Bouwer (1990), who used conservation of mass to derive Equation 7 for linear sorption. The mass-balance procedure uses the flushing factor of Appelo and Postma (1993) written for the entire front. The aim of this paper is to bring this simple method to the attention of hydrochemists.

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