

RATIONAL POLYNOMIAL FUNCTIONS FOR MODELING *E. COLI* AND BROMIDE BREAKTHROUGH

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ABSTRACT. *Fecal bacteria peak concentrations and breakthrough times as affected by preferential flow to subsurface (tile) drainage systems following irrigation or rainfall are important when assessing the risk of contamination. Process-based, convective-dispersive modeling of microbial transport through preferential flow has been conducted. Likewise, regression modeling has been used to study solute transport (e.g., nitrate) under agricultural systems and can have advantages over process-based modeling, such as fewer or easier to determine parameters and easier determination of confidence intervals. However, empirical models (e.g., regression) have only rarely been used to investigate microbial transport. In addition, the selection of time response curves to empirically model simple, right skewed, single breakthrough events from field or laboratory data is generally an arbitrary choice and often considers only conventional distribution-shaped response curves, such as lognormal distributions. In this study, we evaluate four rational polynomial functions for modeling bromide and E. coli data from a single breakthrough event from a tile-drained field near Nashua, Iowa. Bromide and liquid swine manure were applied to the plot immediately prior to 42 mm of overhead sprinkler irrigation. E. coli and bromide concentrations were determined in subsurface drainage water samples collected for the next 24 h. Nonlinear iteratively re-weighted least squares regression procedures were used to model the breakthrough data. The maximum event value, time of occurrence, and event total were estimated from the parameters for each model. Selection of the best model was based on multiple performance criteria. A simple rational polynomial with a linear factor in the numerator and quadratic form in the denominator was the overall best choice for E. coli ($R^2 = 0.92$). A related fractional order form also known as the Gurney model was the best choice for bromide ($R^2 = 0.93$). In comparison, the more commonly assumed lognormal distribution described only 78% of the variation in E. coli and 68% of the variation in bromide, with a weighted mean square error 3.0 to 4.6 times larger than each selected rational polynomial model. In this experiment, the chosen models clearly tracked E. coli and bromide distribution better than the lognormal model.*

Keywords. *Breakthrough curves, Contaminant transport, Empirical modeling, Microbial transport, Subsurface drainage.*

Preferential flow through soil macropores may result in rapid flow and transport to deeper soils and tile drainage systems, including transport of *Escherichia coli* (*E. coli*) (e.g., Abu-Ashour et al., 1994; Christiansen et al., 2004; Akay and Fox, 2007;

Guzman et al., 2009). Understanding and modeling fecal bacteria peak concentration and breakthrough time in tile drainage systems following irrigation or rainfall are fundamental when assessing the risk of contamination from the soil surface to deeper soils and shallow aquifers (McMahon and Christy, 2000; McGechan and Vinten, 2003; Guzman et al., 2009). Developing empirical models and characterizing selected features such as peak concentration and breakthrough time within a confidence interval will help facilitate analysis of *E. coli* transport. Most studies have used the convective-dispersive equation for modeling breakthrough curves of microbial organisms (e.g., Pachepsky et al., 2006; Darnault et al., 2004; Jiang et al., 2007). Thoroughly tested mechanistic models can be transferred to different sites and can complement field and laboratory experiments. For example, models can predict contaminant fate under different conditions where field data are not available or help understand the processes involved, such as preferential flow. Practical considerations, such as difficulty in determination of input parameters or confidence intervals, limit or exclude the use of mechanistic models under some experimental conditions or objectives. Empirically based rational polynomial models (Jaynes et al., 2004) and segmented (splines) polynomial models (Jaynes et al., 2001) have been used to investigate nitrate loss in fields

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under different treatments. However, little information is available on using empirical models (e.g., regression) to analyze *E. coli* breakthrough patterns in tile drainage systems subject to preferential flow.

Single breakthrough events are often characterized by a rapid rise in the constituent of interest from a zero or low level to a single maximum value, and a decline for the remaining time. This behavior is characteristic of many well-known positively (right) skewed probability density functions (PDF), such as those for gamma, Gumbel, and lognormal distributions (e.g., Evans et al., 2000). Relevantly, lognormal distributions are commonly used in many sciences (e.g., Limpert et al., 2001). Specifically, they have been used to model reactive and non-reactive solute transport through soil (e.g., Jury, 1982; Jalali and Rowell, 2008; Stewart and Loague, 2003; Branger et al., 2008). However, breakthrough event data can be more markedly peaked and tail off more slowly than these classical functions. Alternatives to commonly used PDF models include rational polynomials, which have been used to model various phenomena including chemical isotherms (Ratowsky, 1990) and soil phosphorus sorption (Gunary, 1970; Ratowsky, 1986). The parameterizations are simple and straight-forward, initial values are easy to estimate, they may more accurately track breakthrough data, and well-fitting models are less likely to have time series issues, such as autocorrelation of the residuals. Using bromide and *E. coli* data from a common event, we evaluated four related rational polynomial functions and compared them to a lognormal function for modeling breakthrough responses.

MATERIALS AND METHODS

EXPERIMENT AND DATA SELECTION

The field experiment was conducted at Iowa State University's Northeast Research Center near Nashua, Iowa. The study site has a total of 36 plots (58.5 m × 67 m) with soils belonging to the Kenyon-Clyde-Floyd association. A subsurface drainage system discharging to individual wells is located in the center of each plot at 1.2 m depth with tile spacing of 28.5 m (Kanwar et al., 1997, 1999). Manure, bromide, and sprinkler irrigation were applied in sequence to a 30.5 m × 30.5 m area centered above the subsurface drain on the downslope end of plot 30 on 16 April 2008.

Liquid swine manure obtained from a nearby swine finishing facility was injected at the rate of 168 kg N ha⁻¹ parallel to plant rows at approximately 25 cm below the soil surface. Bromide (KBr) was then surface applied as a tracer in granular form at 215 kg ha⁻¹ to the subplot area. Next, water was applied at a rate of 56 mm h⁻¹ (stationary condition) to the experimental plot using a boom, linear-move irrigation system (Valley 6000 pivot, Valmont Industries, Inc., Valley, Neb.) moving 29 m h⁻¹ for two passes. Approximately 42 mm was applied over nearly 2 h. Nozzles (3TN#44, Nelson Irrigation Corp., Walla Walla, Wash.) were assembled vertically downward every 3 m along the boom.

Subsurface drainage water samples were taken for *E. coli* quantification and bromide concentrations at 15 min

increments for the first 3 h of irrigation, every hour until 6 h after irrigation initialization, and then every 3 to 6 h for 24 h total sampling time. Water samples for *E. coli* enumeration were refrigerated and analyzed within 8 h of collection using IDEXX Colilert reagent and the semi-automated QuantiTray 2000 (IDEXX Laboratories, Inc., Westbrook, Maine) method based on the most probable number (MPN) technique, which counts from 1 to 2,419.6 MPN per 100 mL (Garbrecht et al., 2009; Guzman et al., 2010). Water samples for bromide analysis were stored at 4°C until analyzed using the 4500-Br-C method (APHA, 1998).

Bimodal breakthrough patterns were noted for each constituent, possibly due to the two irrigation passes. In addition, we hypothesized that distinctive preferential flow paths and different ratios of matrix flow to preferential flow with faster and slower travel may have contributed to the two peaks. Such an assessment, however, requires further investigation and is beyond the scope and needs of this technical note. The first breakthrough pattern was relatively small and brief (1.5 h) and was excluded for simplicity. Time was reset to zero for the initiation of the second breakthrough, which occurred 2.7 h after irrigation initiation for *E. coli* and at 2.8 h for bromide.

MODEL DESCRIPTION

Table 1 summarizes the rational polynomial models we used. The first model (eq. 1) is a rational polynomial with a first-order polynomial in the numerator and a quadratic form in the denominator. If c were 0, equation 1 would become a form of the well-known Michaelis-Menton model. For equations 1 to 4, time (t) ≥ 0 is the domain of interest. For brevity, let $q = 4ac - b^2$ and $R(t) = a + bt + ct^2$. Initial values for nonlinear regression can generally be obtained by fitting a simple unweighted parabola, $R(t)$, to the transformed dependent variable $t/y(t)$ using a linear regression procedure (table 1). Alternative ways to estimate initial values will be given in the Results and Discussion section.

The corresponding cumulative distribution function (CDF) of equations 1 to 4 under our conditions of $q > 0$ ($Y(t)$; table 1) are obtained by integrating $y(t)$ over time, or $Y(t)$ can be found in published tables (e.g., Gradshteyn and Ryzhik, 1980). Cases of $q \leq 0$ did not occur in our results.

For equations 2 to 4 (table 1), the fractional basis functions are integer powers of $\sqrt[t]{t}$ instead of t . Hence, for equation 2, $R(t)$ becomes $R(\sqrt[t]{t}) = a + b\sqrt[t]{t} + ct$. Equation 2 has 0 power on the highest-order term in the numerator. Equation 3 is a rational fractional polynomial with a first-order fractional polynomial in the numerator and a quadratic form in the denominator. Equation 4 is a rational fractional polynomial with a second-order fractional polynomial in the numerator and a quadratic form in the denominator. Equation 4 is also known as the Gunary model (Gunary, 1970; Ratowsky, 1986, 1990). Initial parameter estimates for nonlinear regression can be obtained by fitting the simple unweighted quadratic form, $R(\sqrt[t]{t})$, to each transformed dependent variable shown in table 1 using a quadratic regression procedure.

Table 1. Rational polynomial models.^[a]

Eq.	Rational Polynomials $y(t)$	Cumulative Distribution Function (CDF)	y_{\max}	Transformation for Initial Values
1	$\frac{t}{a + bt + ct^2}$	$Y(t) = \frac{1}{2c} \log[R(t)] - \frac{b}{c\sqrt{q}} \arctan\left(\frac{b + 2ct}{\sqrt{q}}\right) + const$	$t_{y_{\max}} = \frac{y_{\max}}{(a/c)^{1/2}}$ $a > 0, c > 0$	$\frac{t}{y(t)} = a + bt + ct^2$
2	$\frac{1}{a + b\sqrt{t} + ct}$	$Y(t) = \frac{1}{c} \log[R(\sqrt{t})] - \frac{2b}{c\sqrt{q}} \arctan\left(\frac{b + 2c\sqrt{t}}{\sqrt{q}}\right) + const$	$t_{y_{\max}} = \frac{y_{\max}}{(-b/(2c))^2}$ $c > 0, b < 0$	$\frac{1}{y(\sqrt{t})} = a + b\sqrt{t} + ct$
3	$\frac{\sqrt{t}}{a + b\sqrt{t} + ct}$	$Y(t) = \frac{2}{c} \sqrt{t} - \frac{b}{c^2} \log[R(\sqrt{t})] + \frac{2(b^2 - 2ac)}{c^2\sqrt{q}} \arctan\left(\frac{b + 2c\sqrt{t}}{\sqrt{q}}\right) + const$	$t_{y_{\max}} = \frac{y_{\max}}{a/c}$ $a > 0, c > 0$	$\frac{\sqrt{t}}{y(\sqrt{t})} = a + b\sqrt{t} + ct$
4	$\frac{t}{a + b\sqrt{t} + ct}$	$Y(t) = \frac{t}{c} - \frac{2b}{c^2} \sqrt{t} + \frac{b^2 - ac}{c^3} \log[R(\sqrt{t})] - \frac{2b(b^2 - 3ac)}{c^3\sqrt{q}} \arctan\left(\frac{b + 2c\sqrt{t}}{\sqrt{q}}\right) + const$	$t_{y_{\max}} = \frac{y_{\max}}{(-2a/b)^2}$ $a > 0, b < 0$	$\frac{t}{y(\sqrt{t})} = a + b\sqrt{t} + ct$

^[a] a , b , and c are fitted model parameters; $y(t)$ is the response variable (MPN per 100 mL for *E. coli*, mg L⁻¹ for bromide); t is time (h); q , $R(t)$, and CDF are described in the text; $t_{y_{\max}}$ is the time when $y(t)$ is at its maximum value (y_{\max}); and $const$ represents the constant added to the indefinite integral.

ANALYSIS

Unweighted models were examined first, but diagnostics revealed non-constant error structures (heteroscedasticity). Here, larger estimates were associated with larger residual errors. Consequently, some common inverse variance weights (i.e., weight = σ^{-2}) were considered. Here, weight forms with $\sigma^2 = \hat{y}^p$ with p to be determined were examined. An iteratively reweighted nonlinear least squares procedure was used to develop each model (e.g., Seber and Wild, 1989). Initial conditions for each model were first estimated by using the recommendations provided above. In each case, based on several considerations including residual analyses and dependent variable distributional considerations, the variance model $\sigma^2 = \hat{y}(t)$ was judged to be adequate, where $\hat{y}(t)$ is the predicted value from the given model. Model performance was evaluated based on multiple criteria, including several coefficients of determination, mean square error, several information criteria, autocorrelation, and other standard diagnostics. Some of these tests are based on weighted sum of squares or weighted predictive residual sum of squares (PRESS). For brevity, however, only the coefficient of determination (R^2), the coefficient of determination with PRESS (R_p^2), weighted mean square error (MSE), and Akaike’s information criterion-corrected (AIC_c) are reported. Regression analysis was conducted using PROC NLIN in SAS (ver. 9.2, SAS Institute, Inc., Cary, N.C.). A 95% confidence interval was estimated for each maximum. An inverse regression estimate for the time of each maximum was obtained using the estimate statement in SAS PROC Model, a procedure in the SAS/ETS time series package (SAS ver. 9.2).

Other possible right skewed curves from the exponential family of distributions were considered for the purpose of comparison. These choices included gamma, Gumbel, logistic, lognormal, Raleigh, and Weibull (e.g., Evans et al., 2000). For simplicity, we report only the rational polynomial model results compared to those from the lognormal model (eq. 5), which was the best of these forms for both *E. coli* and bromide series:

$$y(t) = \frac{a}{tb\sqrt{2\pi}} \exp\left\{-\frac{\left[\log\left(\frac{t}{c}\right)\right]^2}{2b^2}\right\} \tag{5}$$

with a , b , and $c > 0$.

Trapezoidal rule integration was used to evaluate and compare the CDF estimates. Note that each total CDF (here in concentration-duration units) is only an estimate because the actual values are unknown, and there are possible problems with either the numerical integration or the analytic integration due to the uncertainty of the parameter estimates and/or the model. In practice, for concentration data like these series, a researcher may want first to convert them to load-rate units to estimate a total load.

RESULTS AND DISCUSSION

For both data sets, all regressions converged, gave parameter estimates within acceptable limits, and had $q > 0$ (table 2). Equation 1 is the best choice for the *E. coli* data ($R^2 = 0.92$; table 2 and fig. 1). For comparison, the lognormal was the best fit of the six classical distributions considered but only had $R^2 = 0.78$ (fig. 1). The Gunary model (eq. 4) is clearly the best for bromide ($R^2 = 0.93$; table 2 and fig. 2). Again for comparison, the corresponding lognormal had $R^2 = 0.68$ (fig. 2). For both selected models, diagnostics revealed no significant autocorrelation in the residuals or other serious problems. Table 3 presents the estimated breakthrough curve peak values (y_{\max}) and their time of occurrence ($t_{y_{\max}}$), with standard uncertainty estimates. All exclude zero and so are distinct. The variance ratios for mean square error values of equation 5 to those of the selected models were 2.99 for the *E. coli* series and 4.60 for the Br series. Each ratio is a significant improvement in interpolation by most statistical and practical criteria.

The totals from the CDF models are listed in table 3. For each data set, the selected model gave a value closest to the numerical integration. There are, however, drawbacks to

Table 2. Breakthrough curve regression results.^[a]

	Equation	R ²	R _p ²	MSE	AIC _c	$a \pm se_a$	$b \pm se_b$	$c \pm se_c$
<i>E. coli</i>	1	0.92	0.84	68	125	$(2.2 \pm 0.4) \times 10^{-3}$	$(-3.1 \pm 0.6) \times 10^{-3}$	$(1.4 \pm 0.2) \times 10^{-3}$
	2	0.90	0.79	90	129	$(7.0 \pm 1.4) \times 10^{-3}$	$(-1.1 \pm 0.2) \times 10^{-3}$	$(4.9 \pm 1.0) \times 10^{-3}$
	3	0.91	0.82	75	126	$(8.5 \pm 1.6) \times 10^{-3}$	$(-1.5 \pm 0.3) \times 10^{-3}$	$(6.9 \pm 1.2) \times 10^{-3}$
	4	0.83	0.73	138	137	$(1.3 \pm 0.3) \times 10^{-2}$	$(-2.3 \pm 0.5) \times 10^{-2}$	$(1.1 \pm 0.2) \times 10^{-2}$
	5 ^[b]	0.78	0.72	205	143	5197 ± 775	0.68 ± 0.072	2.0 ± 0.23
Bromide	1	0.73	0.60	0.41	38	0.11 ± 0.04	-0.011 ± 0.404	0.040 ± 0.009
	2	0.50	0.22	0.82	49	0.31 ± 0.10	-0.24 ± 0.10	0.084 ± 0.038
	3	0.77	0.64	0.35	35	0.41 ± 0.10	-0.53 ± 0.15	0.26 ± 0.05
	4	0.93	0.88	0.11	16	0.76 ± 0.09	-1.34 ± 0.14	0.70 ± 0.06
	5 ^[b]	0.68	0.52	0.52	42	91 ± 20	1.26 ± 0.14	2.2 ± 0.3

^[a] R² is the coefficient of determination; R_p² is the coefficient of determination with the predictive residual sum of squares (PRESS); MSE is the weighted mean square error; AIC_c is Akaike's information criterion-corrected; and $a \pm se_a$, $b \pm se_b$, and $c \pm se_c$ are the regression parameters with their standard errors.

^[b] The parameters for the lognormal model are reported, but they are on a different scale and have no logical correspondence to those in the rational polynomial models.

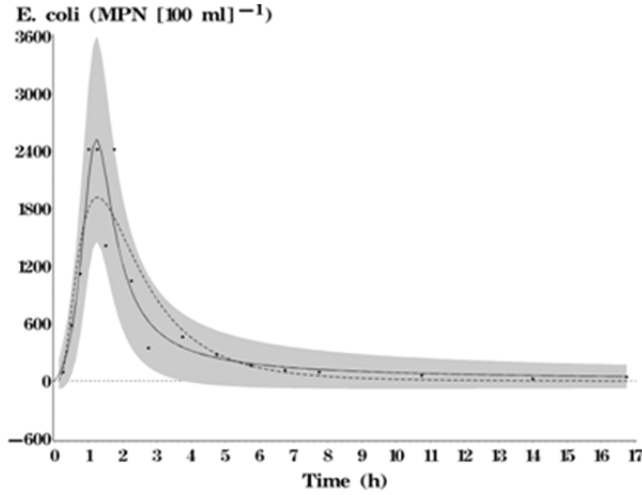


Figure 1. Equation 1 model for *E. coli* breakthrough response: black dots are measured data, dashed line interpolates lognormal model for comparison, solid line interpolates weighted equation 1 regression predictions ($\hat{y}(t)$), and gray band is corresponding 95% confidence interval for $\hat{y}(t)$. Regression results are summarized in table 2.

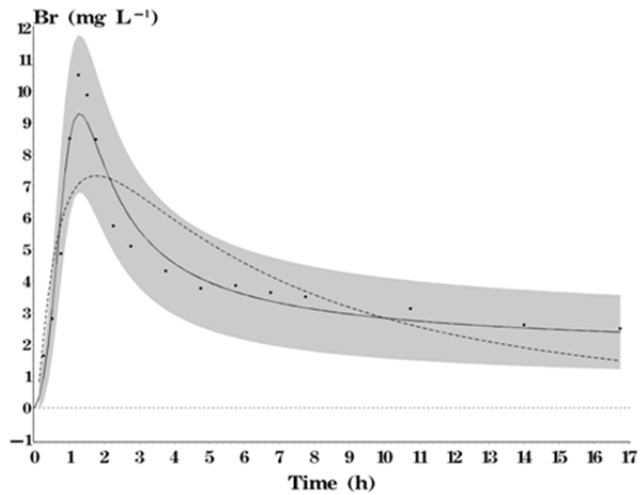


Figure 2. Equation 4 model for the Br breakthrough response: black dots are measured data, dashed line interpolates lognormal model for comparison, solid line interpolates weighted equation 4 regression predictions ($\hat{y}(t)$), and gray band is corresponding 95% confidence interval for $\hat{y}(t)$. Regression results are summarized in table 2.

rational polynomial models. For many of the classical distribution functions, the time of peak value is one of the parameters and so is directly estimated along with its uncertainty. For rational polynomials, this point is a function of the parameters and must be subsequently estimated along with its uncertainty using inverse regression procedures. Finally, as with common PDF models, a weighted regression is usually needed.

Many properties of these curves and comparison of them are of interest. As stated above, each $y_{\max} > 0$ and $t_{y_{\max}} > 0$ in table 3. Other comparisons are likely of more interest to researchers. For example, both bromide and *E. coli* are from the same event, so a question is: do the two $t_{y_{\max}}$ values differ (1.23 h vs. 1.28 h, table 3)? Comparing the confidence limits, obviously not. In the case of different treatments, possible questions for a given constituent are:

Table 3. Breakthrough curve peaks and total estimates.

	Equation	$t_{y_{\max}}$ ^[a]	$l95_{t_{y_{\max}}}$	$u95_{t_{y_{\max}}}$	y_{\max} ^[b]	$l95_{y_{\max}}$	$u95_{y_{\max}}$	Total ^[c]
<i>E. coli</i>	1	1.23 ± 0.06	1.10	1.35	2522	1450	3593	5310 (1.02)
	2	1.35 ± 0.08	1.18	1.51	2374	1203	3545	4912 (0.94)
	3	1.23 ± 0.06	1.10	1.35	2565	1423	3704	5442 (1.05)
	4	1.16 ± 0.06	1.02	1.29	2880	1169	4592	6475 (1.24)
Bromide	1	1.67 ± 0.20	1.23	2.10	8.04	3.77	12.31	59.96 (0.95)
	2	2.05 ± 0.61	0.73	3.36	7.00	1.44	12.56	61.14 (0.97)
	3	1.61 ± 0.18	1.22	1.99	8.29	4.27	12.32	59.19 (0.94)
	4	1.28 ± 0.06	1.15	1.42	9.30	6.81	11.78	61.78 (0.98)

^[a] $t_{y_{\max}}$ is the time for the concentration peak (in hours, with standard error); $l95_{t_{y_{\max}}}$ and $u95_{t_{y_{\max}}}$ are, respectively, the lower and upper 95% confidence interval limits for $t_{y_{\max}}$.

^[b] y_{\max} is the concentration peak value at $t_{y_{\max}}$ (*E. coli* in MPN per 100 mL and bromide in mg L⁻¹); $l95_{y_{\max}}$ and $u95_{y_{\max}}$ are, respectively, the lower and upper 95% confidence interval limits for y_{\max} .

^[c] Total is the value of the model's definite integral (in concentration-duration units). Values in parentheses are ratios of the total to that from a trapezoidal rule numerical integration (5201 for *E. coli* and 62.98 for bromide).

does one treatment delay $t_{y_{\max}}$ or attenuate y_{\max} ?

Many problems can be encountered when using nonlinear regression procedures. Among them are convergence of the iterative procedure and finding reasonable initial values. If convergence is a problem, then: allow for more iterations; loosen the convergence criterion; try a different estimation method; try the CDF model form, other parameterizations, or a closely related model; change the weights; or change the initial values. Each of these suggestions is a subject unto itself. For example, some alternatives for changing the initial values include: try those from an unweighted model first; use those from a regression on the CDF; or try the values from regression results on a closely related data set or model. In this work, however, the procedure suggested in the Materials and Methods section worked the first time in all but one case.

A broader use of these curve forms is possible. For example, modeling multiple or complex breakthrough events with these forms may be possible when the response appears to be a series of single events. Two individual events can be put together by forcing continuity through a common join point (i.e., a knot). Splines or segmented regression are common names for this method (e.g., Seber and Wild, 1989). Additionally, when the data are from a designed experiment with replicated treatments, parameters, peak values, and time of the peak values derived from the response curves parameters may be examined with analysis of variance procedures (e.g., Mead, 1990).

Some applications of low-order rational polynomials are used as purely empirical models because they fit the data well, as was demonstrated in this study. Others, such as isotherm models or sorption processes, can make a mechanistic case for their use. In either case, model design methods should be employed to optimally sample the time response in order to get the best modeling results.

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