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Effects of pollutant speciation in treatment wetlands design

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Abstract

This paper reports on observed and potential effects of distributions of detention times and first order removal rate constants. Rate constants are distributed across the species that make up a grouped class of contaminants, such as total suspended solids (TSS) and biochemical oxygen demand (BOD). The existence of these distributions is shown to invalidate the plug flow (PF) model assumption in almost all cases. The tanks-in-series (TIS) model is shown to offer a better platform to accommodate distributed parameters. Detention time distributions (DTDs) and k-value distributions (kVDs) are shown to both lead to TIS models. Discrete, uniform and gamma distributions for detention and k-values are explored. The greater the variance, the more important the impacts on performance modeling. Data for the reduction of TSS are analyzed from the perspective of particle size distributions, and shown to result in the appearance of TIS behavior for the lumped TSS measure. The retarded rate constant concept is shown to be subsumed by the idea of a weathering mixture with a distribution of rate constants.

Keywords: Constructed wetlands; Distributions; Detention time; Mixtures; Lumped parameters; Depth; Suspended solids

1. Introduction

Most of the treatment wetland literature utilizes univariate analyses, in the sense that single numbers are used for items such as detention time, concentration, and rate constants. However, many treatment wetland variables and parameters do not possess single unique values, but are distributed with respect to some wetland attribute. Two such distributed variables are explored here: detention time and first order rate constants.

The most studied of these is the detention time distribution (DTD), which characterizes the travel times of water elements leaving the wetland (Kadlec, 1994; Kadlec and Knight, 1996). Elements of water traverse the wetland in different times, either because they follow different parallel paths or because of dispersive mixing. In either case, there exists a probability distribution of transit times, which is virtually always a delayed, skewed bell-shaped curve. A variety of mechanistic models have been utilized to describe wetland DTDs: tanks in series, plug flow (PF) with dispersion (Kadlec and Knight, 1996), finite and infinite stage (Mangelson, 1972; Werner and Kadlec, 1999). The end result of all experiments and models is the prediction of extreme sensitivity of high levels of pollutant reduction to the character of the DTD (Levenspiel, 1972).

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In almost all instances, water quality parameters are measured by procedures that lump individual chemical compounds into an overall, or total concentration for that class of materials. Biochemical oxygen demand (BOD) and total suspended solids (TSS) are examples of such lumping. It is clear that the individual components of such mixtures may be degraded or removed at different rates, and that there is a corresponding difference in removal rate constants (Tchobanoglous et al., 2000; Crites and Tchobanoglous, 1998). There is, therefore, a distribution of rate constants across the various mass fractions of the mixture. Such a distribution may be discrete, for the case of a countable and very small number of individual compounds; or it may be continuous, for the case of a very large and possibly uncountable number of constituents. Combinations of both types of distribution are also common, such as for total nitrogen (TN). TN consists of a few separately identifiable compounds (nitrate, ammonia) plus lumped classes of compounds (particulate N, organic N). Total phosphorus (TP) is comprised of particulate (PP), and dissolved organic (DOP) and soluble reactive (SRP) forms. As water containing such a mixture passes through the wetland, its composition changes because different fractions of the mixture are reduced at different rates. The mixture becomes ‘weathered,’ a term coined to describe the selective stripping of light volatile materials upon exposure to outdoor environments.

The objective of this paper is to illustrate the potential effects of these two types of parameter distributions on the behavior of wetlands for pollutant reduction. Observations are typically made of removal versus either mean detention time (frequent) or distance through the wetland (less frequent). It will be shown that the existence and character of parameter distributions can have strong influences on performance.

The basis for this investigation is the presumption of a first order removal model, with a 0 background concentration. However, extension of the results to the case of a non-zero background is easily done, by replacing fractions remaining with fractions remaining to background. First order models may be either area-based or volume-based (Kadlec and Knight, 1996). These are equivalent for constant depth systems, with the volumetric rate constant equaling the area-based rate constant divided by the water depth. In this paper, the volumetric rate constants are utilized, except for particle settling, which is more intuitive on an area basis.

2. Distribution mathematics

Three different types of distribution functions will be used: discrete, linear and gamma distributions.

A discrete distribution function is defined by:

$$f_i = f(x_i) \quad i = 1,\ 2,\ 3,\ \ldots,\ N \tag{1}$$

The normalized form is assumed, that is:

$$\sum_{i=1}^{N} f_i = 1 \tag{2}$$

In this formulation, the f_i are the fractions of the population that possess property values x_i. The average value of x is:

$$\bar{x} = \sum_{i=1}^{N} f_i x_i \tag{3}$$

The uniform distribution is:

$$f(x) = \begin{cases} 0, & 0 < x < a \\ \dfrac{1}{(b-a)}, & a < x < b \\ 0, & x > b \end{cases} \tag{4}$$

where, a is lowest x-value; b is highest x-value.

The interpretation of the function $f(x)$ is that it equals the fraction of the population with x-values between x and $x+\mathrm{d}x$. The normalized form is assumed, that is:

$$\int_0^\infty f\ \mathrm{d}x = 1 \tag{5}$$

This distribution represents the case of a finite range of possible x-values, with equal amounts of the population in every sub-range. The mean value of x for this distribution is the average of the upper and lower bounds:

$$\bar{x} = \int_0^\infty xf \, dx = \frac{(a+b)}{2} \tag{6}$$

The uniform distribution is a limiting case of finite linear distributions:

$$f(x) = \begin{cases} 0, & 0 < x < a \\ A + \dfrac{(B-A)(x-a)}{(b-a)}, & a < x < b \\ 0, & x > b \end{cases} \tag{7}$$

The gamma distribution is (Wadsworth et al., 1990):

$$f(x) = \frac{1}{\beta \Gamma(n)} \left(\frac{x}{\beta}\right)^{n-1} \exp\left(-\frac{x}{\beta}\right) \tag{8}$$

where, n; shape parameter, β; scaling parameter, $\Gamma(n)$; Γ function of n,

$$\Gamma(n) = \int_0^\infty x^{n-1} e^{-x} \, dx \tag{9}$$

This distribution is also normalized, that is it integrates to unity. The mean value of x for the gamma distribution is:

$$\bar{x} = n\beta \tag{10}$$

When n is an integer, the Γ function is $\Gamma(n) = (n-1)!$ (a factorial). When $n = 1$, the gamma distribution becomes the exponential distribution. Both the gamma distribution and the Γ function are readily available in handbooks (e.g. Dwight, 1961), or as computer spreadsheet tools (e.g. GAMMADIST and GAMMALN in EXCEL™). The uniform and gamma distributions of x-values are illustrated in Fig. 1.

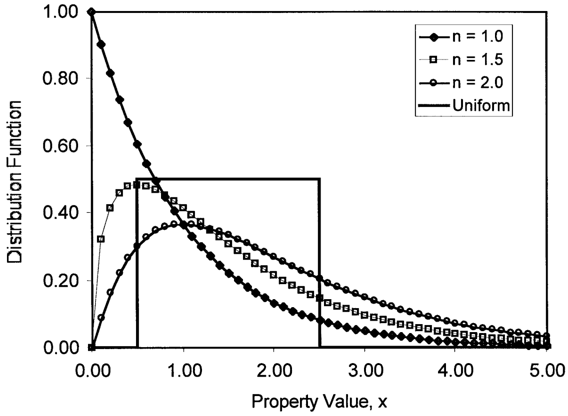

Fig. 1. Shapes of the uniform and gamma distributions. The uniform distribution is illustrated for $a = 0.5$ and $b = 2.5$. The gamma distribution is illustrated for $n = 1.0$, 1.5, and 2.0; and $\beta = 1.0$. The case of $n = 1$ represents the exponential distribution.

3. Detention time distribution functions

Water travels tortuous paths through wetlands, due to several environmental factors. It follows fast and slow tracks, created by topography and patterns of vegetation (Kadlec and Knight, 1996). Wind, waves and submersed vegetation create eddies and mixing in free water surface (FWS) wetlands. In subsurface flow (SSF) wetlands, particulate media cause similar tortuous routing and eddy mixing. The result of these phenomena is a distribution of detention times in the treatment wetland. Importantly, the distribution may result solely from stratified velocity profile effects, without any contribution from mixing processes.

DTDs for treatment wetlands have been extensively investigated at a large number of sites, and thus there exist numerous examples of the functional forms that are characteristic of wetlands (for example: Grismer et al., 2001; King et al., 1997; Dal Cin and Persson, 2000; Stern et al., 2001). Single-shot tracer injection with effluent concentration monitoring is usually employed (Kadlec and Knight, 1996). Examples of distributions are shown in Fig. 2, which are typical of the larger literature database. These distributions may be described by several different model equations, but commonly the tanks-in-series (TIS) model is applied (Levenspiel, 1972; Kadlec, 1994). The result is a gamma distribution with $n = N$ and $\beta = t_i$:

$$g(t) = \frac{1}{t_i (N-1)!} \left(\frac{t}{t_i}\right)^{N-1} \exp\left(-\frac{t}{t_i}\right) \tag{11}$$

where, t is detention time, day; t_i is mean detention time in one tank, day, N is number of tanks.

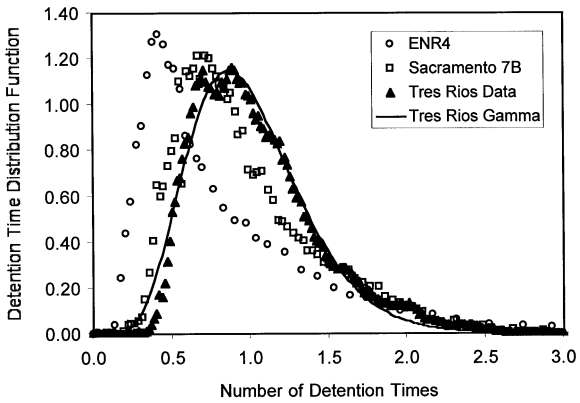

Fig. 2. DTDs for three wetlands. Trés Rios H1 is a 1.3 ha FWS wetland with five cross-ditches and an aspect ration of L:W = 3.8 (Wass, 1997). The line is the gamma distribution model for Trés Rios H1, with $N = 7.4$. Sacramento 7B is a 0.3 ha FWS wetland with three cross-ditches and an aspect ratio of L:W = 12.6 (Dombeck et al., 1998), for which $N = 6$. ENR 4 is a 147 ha FWS wetland, triangular fed from the base, with longitudinal deep channels (DB Environmental, 2000), for which $N = 1.3$.

The mean value of the detention time (τ) is:

$$\tau = Nt_i \tag{12}$$

Eq. (11) may be generalized to a fractional number of 'tanks' by use of the gamma function:

$$g(t) = \frac{1}{t_i \Gamma(N)} \left(\frac{t}{t_i}\right)^{N-1} \exp\left(-\frac{t}{t_i}\right) \tag{13}$$

Thus it is seen that the TIS distributions are gamma distributions, in which the scaling factor is the total detention time divided by N. Eq. (13) may be easily fit to tracer data by selecting N and t_i to minimize error.

It is to be noted that although gamma distributions describe TIS mixing, the converse is not true. A gamma distribution of detention times does not imply the existence of turbulent mixing. Indeed, a gamma distribution may also arise from totally unmixed, separate travel paths with different velocities.

In the limit as N becomes very large, the gamma distribution becomes the PF distribution, with all water departing after exactly one nominal detention time. This limiting case does not exist for treatment wetlands. Virtually all reported literature values fall in the range $1 \le N \le 8$. The question thus arises whether the PF assumption is adequate for performance computations. Comparison computations may be made using the integrated mass balance equations:

$$\text{For plug flow: } \frac{C}{C_\mathrm{i}} = \exp(-kt) \tag{14}$$

$$\text{For } N \text{ TIS: } \frac{C}{C_\mathrm{i}} = \frac{1}{(1 + kt/N)^N} \tag{15}$$

where, C is outlet concentration, gm m^{-3}; C_i is average inlet concentration, gm m^{-3}; k is first order volumetric rate constant, per day.

Graphical methods are available to estimate the effect of non-PF distributions on wetland pollutant removal (Kadlec and Knight, 1996). It is noted that at the 75% removal level, errors in PF removal range from 10% at $N = 8$ to 40% at $N = 1$. Thus the reported N values typically place wetlands in the category of significant departure from PF conditions.

4. *k*-value distribution functions

A lumped constituent in water entering a treatment wetland may possess one of several types of distributions of the species that comprise that lumped constituent. First, the material in question may be either discretely or continuously distributed. An example of a discrete inlet distribution is a mixture of three alcohols: methanol, ethanol and propanol dissolved in water, with total alcohol as the lumped measure of contaminant concentration. The species distribution consists of only three discrete numerical fractions. Each of the species will have its own removal rate, and most often these will be different. It is easy and common practice to deal with this situation by considering the individual species separately, and ultimately summing the species concentrations to obtain the total concentration of the lumped class of compounds.

An example of a continuous inlet distribution is a mixture of solids of different sizes, spanning a continuum from small to large particle sizes. TSS is the lumped measure used exclusively in connection with water quality. The concept of the particle size distribution is well known, and the lognormal distribution of particle numbers across sizes is commonly found. The expression for the size frequency distribution function $f(D)$ is:

$$f(D)\mathrm{d}D = \text{number fraction of particles in size range } D \text{ to } D + \mathrm{d}D \tag{16}$$

where, D is particle diameter, m.

The expression for the cumulative size distribution function $F(D)$ is:

$$F(D) = \text{number fraction of particles of size less than } D \tag{17}$$

These two distributions are related by:

$$F(D) = \int_0^D f(D)\,\mathrm{d}D \tag{18}$$

The rate of particle removal in a wetland is often related to the particle size and density, and so each size category will have a different rate of removal in the wetland.

However, for the present purposes, the number distribution of sizes is not what is needed. Each fraction of the distribution, whether discrete or continuous, will here be presumed (the unhindered settling assumption) to be removed in the wetland according to a first order kinetic model:

$$J = -kC \tag{19}$$

where, C is concentration, gm m^{-3}; J is removal rate, gm m^{-2}·per day; k is areal removal rate constant, m per day.

Each fraction of the lumped material will in general possess its own k-value. Therefore, there is a distribution of k-values, designated by $f(k)$:

$$f(k)\mathrm{d}k = \text{mass fraction of material with rate constant in the range } k \text{ to } k + \mathrm{d}k \tag{20}$$

In the case of the example of particulate matter removal, note that the settling rate is often proportional to the square of the particle diameter, and the mass is proportional to the cube of the particle diameter (ASCE, 1975). Therefore, the mass distribution function for settling rate constants is different than the size distribution. Here the k-value frequency distribution across the mass fractions of the lumped material is termed the k-distribution (kVD).

In the wetland environment, the DVD and the kVD interact to produce the overall observed reduction in a lumped category of pollutants. However, batch testing eliminates the DVD effect, since there is no distribution function for batch time. The DVD effect is also removed in theory for the (unachievable) ideal of true PF.

4.1. Plug flow or batch systems with distributed k-values

As an illustrative case, the hypothetical (non-existent) PF flow pattern is combined with a gamma distribution of k-values. This is important as a reference to the large existing literature based on the faulty PF assumption, and as a descriptor of batch processes in well-mixed environments. Each element of entering water will spend exactly one detention time in the wetland. However, the different portions of the lumped pollutant will undergo different degrees of reduction, according to their respective k-values, and according to Eq. (14). For gamma distributions, the average total pollutant remaining is:

$$\frac{\bar{C}}{C_\mathrm{i}} = \int_0^\infty \mathrm{e}^{-kt} f(k)\,\mathrm{d}k = \int_0^\infty \mathrm{e}^{-kt}\frac{1}{\beta\Gamma(n)}\left(\frac{k}{\beta}\right)^{n-1}\exp\left(-\frac{k}{\beta}\right)\mathrm{d}k \tag{21}$$

where, $\bar{C}$ is average outlet concentration, gm m^{-3}; t is elapsed time, day.

The integration in Eq. (21) produces the result:

$$\frac{\bar{C}}{C_\mathrm{i}} = \frac{1}{(1+\beta t)^n} \tag{22}$$

Using the mean value of k from Eq. (10) for the starting mixture, this result is:

$$\frac{\bar{C}}{C_\mathrm{i}} = \frac{1}{(1+k_0 t/n)^n} \tag{23}$$

where, k_0 is average rate constant for the starting mixture, per day.

It may also be shown (the details are omitted here) that the average k-value at any time during the reduction process is:

$$k = \frac{k_0}{(1 + \beta t)^n} \tag{24}$$

This form has been directly presented in the literature, without the supporting concept of the k-value distribution function (Crites and Tchobanoglous, 1998; Tchobanoglous et al., 2000; Shepherd et al., 2001). Due to the average k-values decrease with travel time in the wetland, or equivalently with batch time, these authors call the concept 'retarded' or 'time dependent' rate constants. However, nothing happens to the component rate constants during passage of time; rather, it is the composition of the mixture that changes.

As weathering proceeds, it may be shown from the mathematics that gamma distributions are preserved. The shape parameter n remains unaffected, but the scaling factor β changes with weathering time. The distribution shifts to favor slower rate constants. Fig. 3 illustrates the evolution of $f(k)$ for a starting mixture with $n = 3$ and $\beta = 0.5$ per day (starting $k_0 = 1.5$ per day).

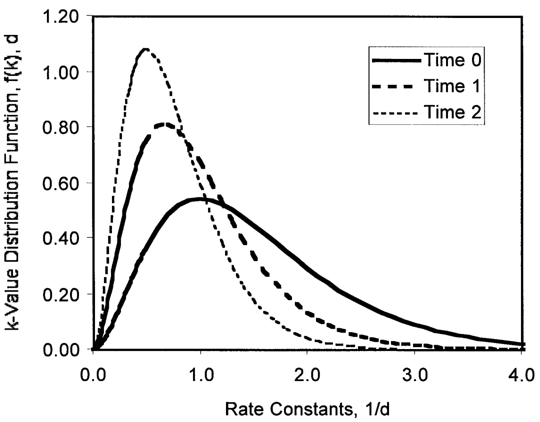

Fig. 3. Evolution of the kVD due to weathering of a contaminant mixture. The gamma distributions all have $n = 3$. The average k for the mixture decreases from 1.5 to 0.75 to 0.5 per day as t increases.

4.2. A generalization: the relaxed TIS model

Eq. (23) is identical in form to Eq. (15), but the parameters k_0 and n represent the k-distribution for incoming materials comprising the lumped pollutant, rather than the distribution of detention times. This similarity in form leads to the concept that different factors, such as either the DTD or the kVD, can lead to similar model forms. It is likely that observed behavior in real wetland situations is well represented by the TIS equation, wherein the parameter values are relaxed to become fitting parameters. The relaxed TIS concentration model is, therefore, here defined to be:

$$\frac{C}{C_i} = \frac{1}{(1 + k_{app} t/P)^P} \tag{25}$$

where, k_{app} is apparent TIS rate constant, per day; P is apparent number of TIS.

4.3. An example: batch wetland TSS reduction

Suspended particulate matter is removed from water by settling, which may be described by a first order model for each size fraction. In this example, a quiescent batch settling experiment is considered, for which there is no resuspension. The lumped measure of this water contaminant is TSS. Water containing TSS was obtained from the Houghton Lake, MI treatment wetland, and subjected to laboratory analysis. This FWS wetland has been described in the literature (Kadlec and Knight, 1996). High-resolution optical microscopy was used to determine the size distribution, which is the number density across size (Fig. 4). However, the distribution of interest here is not the size distribution; it is the mass distribution of settling velocities, which are the first order areal removal rate constants. In general, settling velocities are proportional to the square of particle size, with variation including shape factors and particle density. Particle mass was estimated to be roughly proportional to the cube of size. These relations allow the conversion of the size distribution to a settling velocity distribution (Fig. 5). That kVD is a uniform distribution (Eq. (4)), with $a = 0$ and $b = 0.34\ \mathrm{min}^{-1}$ ($R^2 = 0.94$). Integration provides

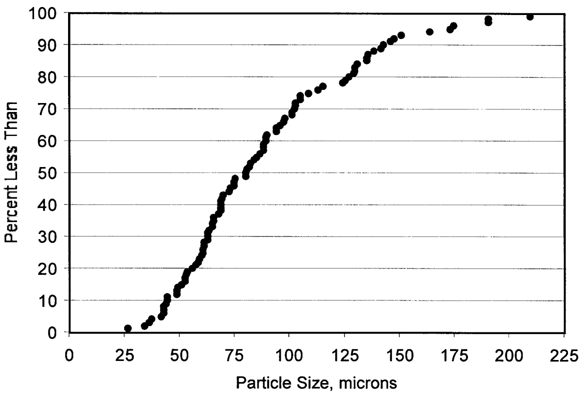

Fig. 4. Cumulative size distribution for Houghton Lake TSS.

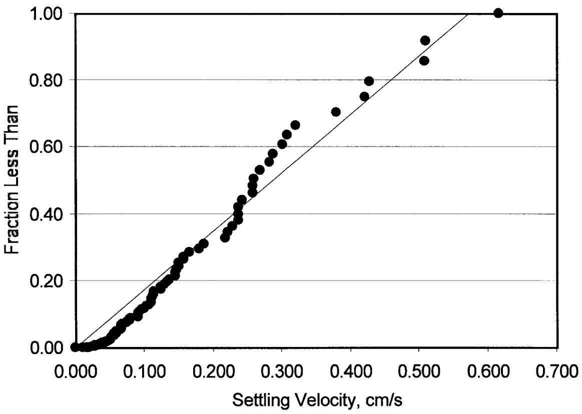

Fig. 5. Cumulative mass distribution of settling velocities for Houghton Lake TSS. This cumulative distribution is a straight line of slope = 0.57 s cm^{-1} ($R^2 = 0.94$). The associated density distribution is $f(k) = 1.75$, $0 \leq k \leq 0.57$ cm s^{-1}.

the fraction remaining:

$$\frac{\bar{C}}{C_i} = \int_0^{\infty} e^{-kt} f(k) \mathrm{d}k = \int_0^{b} e^{-k\tau} \frac{1}{b} \, \mathrm{d}k = \frac{(1 - e^{-bt})}{bt} \tag{26}$$

Batch column settling experiments were also conducted, using the same wetland TSS, thus allowing an independent exploration of the settling distribution. A well-mixed quantity of water and TSS was placed in the column, and the mix sampled for TSS at a succession of times thereafter. Eq. (23) fits experimental settling column data very well when the constant b is determined from that data (Fig. 6). However, the fit may also be accomplished by converting the particle size distribution, as detailed above, with one degree of freedom associated with the shape and density factors. That adjustment is a simple time scale factor. This fit derived from the initial particle size distribution is also reasonable (Fig. 6).

The mass settling data may also be fit with a relaxed PTIS model (Fig. 6). This empirical approach provides an excellent fit ($R^2 = 0.998$). However, the apparent number of TIS is $P = 0.80$, which gives the appearance of being very much removed from the PF mixing conditions associated with the batch test.

The mean settling rate constant decreases as the mixture weathers. Large particles settle, leaving a mixture of smaller particles that settle more slowly. The initial uniform distribution, which is a truncated limiting case of the exponential distribution, is transformed to steeper, truncated exponential distributions. Therefore, while Eq. (24) is derived for gamma distributions, and is therefore not strictly applicable, it nevertheless provides an extremely good fit for the mean k-values as a function of time. For the Houghton Lake example TSS distribution, $k_0 = 0.285$, $n = 2.15$ and $\beta = 0.059$.

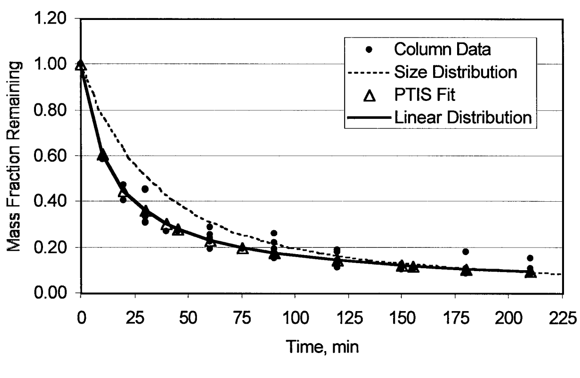

Fig. 6. Reduction of Houghton Lake TSS as a function of time in batch settling tests. Data points are from nine lab experiments. The solid line is the Eq. (25) fit. The dashed line is the fit using the size distribution and calibrated for the density/shape factor. The triangles represent a relaxed PTIS fit (Eq. (26)), with $R^2 = 0.998$, $P = 0.80$ and $k = 4.17$ h^{-1}.

4.4. The example extended: batch wetland reduction of solid-partitioned substances

Many pollutants, such as metals, nutrients and organics partition to, or sorb to, suspended particulate matter. Consequently, removal of TSS implies removal of the sorbed fraction of those materials. This means that distributions of removal rates are again involved in the overall observed performance. Partitioned materials are often proportional to the surface area of a particle. That means that small particles, with large specific surface area, contain relatively large amounts of sorbed substances, compared with large particles. If it is presumed that sorbed contaminant mass is proportional to particle surface area, then the distribution of interest is the particle area fraction distributed across settling velocities, which again are the first order removal rate constants.

When the uniform distribution of particle masses across settling velocities (Eq. (8)) for the Houghton Lake example is converted to the distribution of particle areas across settling velocities, a decreasing linear distribution results:

$$f_A(k) = \begin{cases} \dfrac{2(b-k)}{b^2} & 0 < k < b \\ 0 \quad k > b \end{cases} \tag{27}$$

where, b is highest k-value, $b = 0.57$ per day; $f_A(k)$ is areal distribution function = sorbed mass distribution function, day.

This function reflects the fact that tiny particles, with large area and hence large sorbed pollutant mass, settle very slowly; while large particles carry little pollutant and settle rapidly. The function (Eq. (27)) is valid for the initial particle mixture, but the mixture weathers as settling proceeds. The decreasing linear function trends toward an exponential distribution (not demonstrated here).

It is informative to consider the fractional removal of the sorbed pollutant as a function of the batch settling time. An integration similar to that indicated in (Eq. (21)) produces the fraction remaining of the sorbed pollutant at any time t in the batch settling experiment (Fig. 7), which shows a slowing decline in the fraction remaining. This decline may be modeled either with the distribution function technique, or with a '$k-C^*$' model (Kadlec and Knight, 1996):

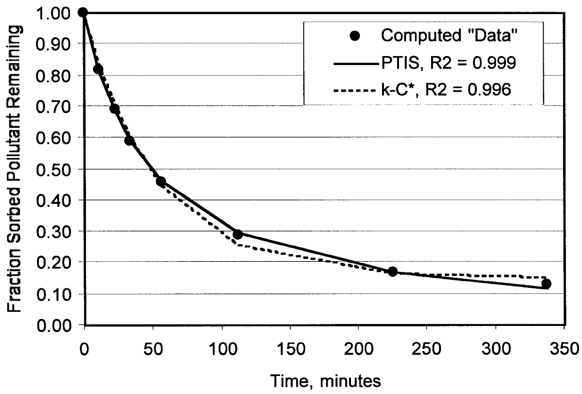

Fig. 7. Reduction of a pollutant partitioned to TSS as a function of time in batch settling tests, based on Houghton Lake sediment size distribution data. Data points are presumptive removals based on area. A posteriori fits via relaxed TIS and $k-C^*$ models are shown. For the TIS model, $P = 1.07$ and $k = 1.20\ \mathrm{h}^{-1}$. For the $k-C^*$ model, $C^* = 0.153C_i$ and $k = 1.13\ \mathrm{h}^{-1}$.

$$\frac{(C - C^*)}{(C_i - C^*)} = \exp(-kt) \tag{28}$$

Both relaxed TIS and $k-C^*$ models fit the pattern very well. However, the effect of the distributed k-values is to create the appearance of a TIS batch time behavior ($P = 1.07$), or to create an apparent $C^* = 0.153C_i$.

5. Combined distributions

Eq. (14) shows that the rate constant and the detention time occur as a product, defined as the Damköhler number, Da $= kt$. The rate constant k may be distributed, and the detention time t is distributed. Thus under any circumstances, the Damköhler number will be distributed. Many experimental studies have determined DTDs that are gamma distributions. There is a very limited knowledge base for kVDs, but the analysis of the preceding section strongly suggests that a relaxed TIS model is applicable to contaminants contained within or adsorbed on TSS. Other compound groups, such as BOD, may also have rate constants distributed by some form of the gamma distribution. Pollutant removal calculations are accomplished for this situation by averaging removals using both of the distribution functions.

5.1. Gamma-distributed flow with gamma-distributed k-values

The real wetland situation is that of a DTD in addition to the kVD presumed here. This leads to the necessity for double averaging to obtain the mean outlet concentration:

$$\frac{\bar{\bar{C}}}{C_{\rm i}} = \int_0^\infty g(t)\left\{\int_0^\infty e^{-kt}f(k){\rm d}k\right\}{\rm d}t = \int_0^\infty \frac{g(t)}{(1+\beta t)^n}{\rm d}t \tag{29}$$

where $g(t)$ is given by Eq. (13). The inner integral is given by (Eq. (23)). Eq. (29) is not amenable to a closed form solution, and therefore the result must be obtained by numerical integration. There are now four parameters to be set: N and t_i for the DTD, and n and β for the kVD. The parameters n and N do not sum to give an effective number of TIS. Indeed, n is a denominator power in (Eq. (29)), while N is a numerator power, as part of $g(t)$. Thus the n and N parameters act in opposite directions.

As an illustration, the parameter $N=3$ is chosen for the DTD, which is typical of many wetland results (Kadlec and Knight, 1996). The parameter $\tau = Nt_i$ is varied, to represent different travel times (distances) through the wetland. The parameters selected for kVD are $\beta = 0.2$ per day and $n = 1.0$, and $k_0 = 0.2$ per day. These are estimated values for BOD from Crites and Tchobanoglous (1998). This example shows a decreasing trend of concentration with detention time as expected (Fig. 8). However, the average k-value decrease causes an apparent asymptote above 0 concentration. The pollutant is not removed to the extent predicted by a constant, inlet k-value, whether or not PF is assumed (Table 1). However, the PF assumption is worse than the PTIS assumption in this regard.

The key point here is that measuring and knowing the DTD does not compensate for knowing the kVD. Even the best fit to the 'data' using three TIS is badly (ca. 50%) in error at the low end of the concentration range. This means that inert tracer studies do not serve to determine the best PTIS model parameters for the case of gamma distributed k-values. The effect in this illustration has been to decrease the number of tanks from 3 to 0.8.

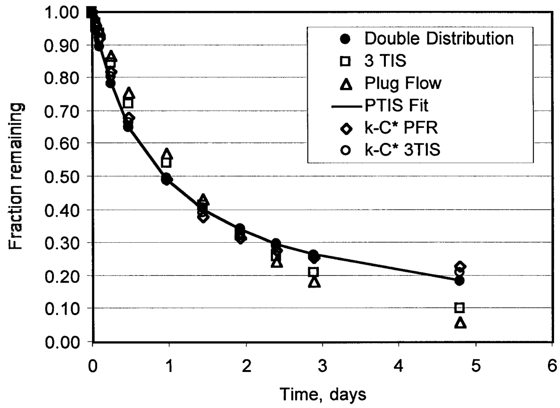

Fig. 8. Lumped pollutant removal in wetlands. The solid points represent the forecast for a three-TIS DTD coupled with a gamma distribution of k-values. The k-distribution parameters were $n = 1.0$, $\beta = 0.2$ per day and $k_0 = 1.25$ per day. Open squares represent a best-fit k value of 0.713 per day coupled with the three TIS DTD. Open triangles represent PF (impulse DTD) with a best-fit k value of 0.584 per day. The line is an a posteriori fit to the PTIS model, in which k and the number of TIS is adjusted. The resulting PTIS parameters were $k = 1.18$ and $P = 0.80$.

5.2. Curve fitting for simultaneous kVD and DVD

The gamma function DTD and the gamma function kVD each separately produce a concentration (Eqs. (15) and (24)) that is of the same form as the TIS model. It is likely that the observed behavior for simultaneous continuous kVD and DVD is well represented by the PTIS equation, wherein the parameter values are relaxed to become fitting parameters (Eq. (24)).

The gamma DTD/kVD model was used to generate concentration profiles for an incoming $k_0 = 0.2$ per day, and ranges $1 \le n \le 8$ and $1.5 \le N \le \infty$. That value of k_0 is appropriate for nutrients, such as TP or TN, which are more difficult to remove than BOD, and hence have

Table 1
Goodness of fit for double continuous distributions

	Set parameters (N)	Best-fit parameters	Sum of squared errors	R^2
PFR	∞	$k=0.584$	0.052	0.941
DTD	3	$k=0.713$	0.025	0.972
PFR $k-C^*$	8	$C^*=0.23$; $k=1.116$	0.007	0.992
3TIS $k-C^*$	3	$C^*=0.18$; $k=1.192$	0.002	0.997
PTIS	–	$P=0.80$; $k=1.180$	0.000	1.000
$k-C^*$	–	$N=0.80$; $C^*=0$; $k=1.180$	0.000	1.000

lower k-values. That computer data was then fit with Eq. (24), generating k_{app} and P values for each profile. The value of the apparent rate constant was constant across all distributions, $k_{\mathrm{app}}=0.198\pm0.006$ (mean $\pm$ S.D.). The apparent number of TIS varied systematically, but always $P\leq N$ and $P\leq n$ (Table 2). The correlation coefficients for these fits all had $R^2=1.000$.

The $k-C^*$ PFR model was also fit to the computer data profiles, with non-zero C^* as a result (Table 3). These 'data' fits were also excellent, with $R^2=0.994\pm0.006$ (mean $\pm$ S.D.). The fit artificially introduces C^* values up to 35% of the incoming concentration. The rate constants are close to, but not equal to, the intrinsic $k_{\mathrm{avg}}=0.2$ per day.

6. Discrete kVD

In contrast to continuously aggregated mixtures, such as TSS and BOD, some pollutants are measured as groups of small numbers of individual compounds.

Table 2
Apparent TIS numbers (P) for hypothetical double continuous distributions

N	$n=1$	$n=4$	$n=8$	$n=\infty$
1.5	0.54	0.99	1.13	1.50
2	0.68	1.37	1.62	2.00
4	0.86	2.19	2.86	4.00
8	0.95	2.90	4.29	8.00
∞	1.00	4.00	8.00	–

6.1. A hypothetical example

Since the discrete distributions have no unifying equation comparable to those for continuous distributions, a hypothetical example is used to illustrate the concepts and effects. A mixture of three species of contaminant is fed to a wetland. The lumped sum of the species is measured in the field, so that:

$$C=C_1+C_2+C_3 \tag{30}$$

Each of these species has a different k-value, assumed to be:

$$k_1=0.08 \text{ per day},\quad k_2=0.12 \text{ per day},\quad k_3=0.40 \text{ per day}$$

(note: for a 30 cm deep FWS wetland, these correspond to areal rate coefficients of $k_{1\mathrm{A}}=8.8$, $k_{2\mathrm{A}}=13.1$, $k_{3\mathrm{A}}=43.8$ m per year). Entering the wetland, there are equal amounts of each of the three species, totaling 100 gm m^{-3}. It is further assumed that the wetland internal hydraulics are described by a three TIS DTD. The three first order disappearances proceed at different rates (Fig. 9), with compound 3 being depleted most rapidly. It is presumed in this example that only the sum of these three species is measured, which follows the trend of the circles in Fig. 10. This hypothetical data may be fit with simple models, either the TIS DTD or the idealized, benchmark PFR distribution.

The solid line in Fig. 11 represents the relaxed PTIS model fit. The representation is very good ($R^2=0.999$), and the apparent rate constant $k_{\mathrm{app}}=0.18$ per day is in the range of individual species k-values. But the apparent number of TIS is $P=1.5$, which is much lower than the hydraulic

Table 3
$k-C^*$ parameters for double continuous distributions

N	$n=1$		$n=4$		$n=8$		$n=100$	
	k	C^*	k	C^*	k	C^*	k	C^*
1.5	0.23	0.35	0.22	0.23	0.22	0.21	0.22	0.18
2	0.20	0.29	0.20	0.16	0.20	0.14	0.20	0.12
4	0.17	0.19	0.18	0.07	0.18	0.05	0.19	0.04
8	0.15	0.13	0.17	0.03	0.18	0.02	0.19	0.01
100	0.10	0.02	0.16	0.00	0.18	0.00	0.20	0.00

Values of C^* are fractions of the inlet concentration.

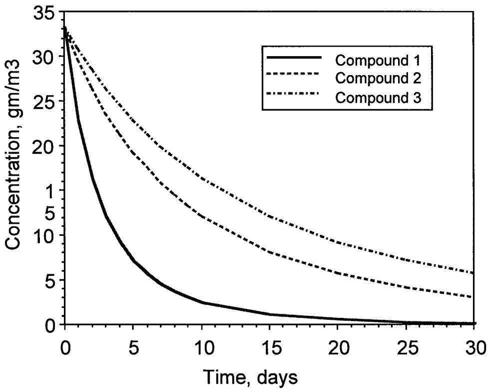

Fig. 9. Concentration reductions for three species with different k-values.

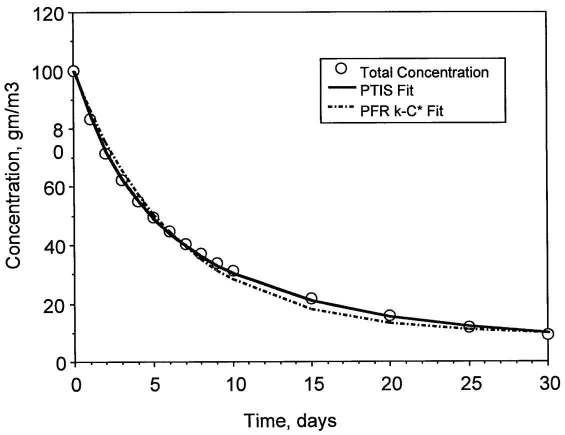

Fig. 10. Computed total concentration of three contaminants of different k-values in a three TIS wetland. The relaxed PTIS fit to this hypothetical data (solid line) has $P=1.5$ and $k_{app}=0.18$ per day. The relaxed PFR $k-C^*$ fit to this hypothetical data (dashed line) has $C^*=9$ gm m^{-3} and $k_2=0.16$ per day.

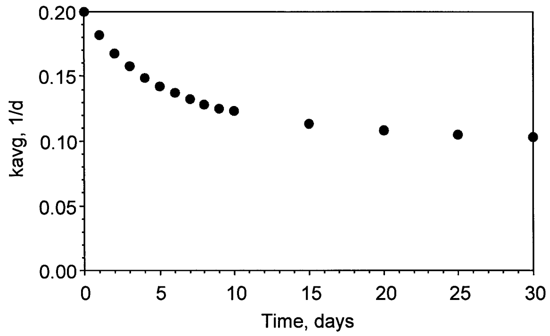

Fig. 11. Mass average k-value in a three component mixture as a function of time.

model value of $N=3$. The discrete distribution of k-values has created the illusion of reduced hydraulic efficiency (fewer TIS).

A second candidate model is the PFR $k-C^*$ model (Kadlec and Knight, 1996). The hypothetical 'data' in Fig. 10 display a trend toward a plateau with increasing detention time, a feature accounted in the PFR $k-C^*$ model:

$$\frac{(C - C^*)}{(C_i - C^*)} = \exp(-k_2 t) \tag{31}$$

where, k_2 is apparent PFR $k-C^*$ rate constant, per day; C^* is apparent plateau concentration, gm m^{-3}.

The dashed line in Fig. 10 represents the relaxed PFR $k-C^*$ model fit. The representation is also very good ($R^2=0.990$), and the apparent rate constant $k_2=0.16$ per day is in the range of individual species k-values. But the apparent background concentration $C^*=9$ gm m^{-3} does not exist in reality. All three constituents eventually are reduced to 0 according to the generation method. The discrete distribution of k-values has created the illusion of a non-zero background concentration.

The mass average k-value for the mixture decreases with time, as the more readily removed component is lost (Fig. 11).

6.2. A wetland example: surfactant reduction

Alkylbenzenesulfonates occur as mixtures of isomers of homologous series of carbon numbers 11–14. Wetlands remove these substances very effectively (Del Bubba et al., 2000). The data of Inaba (1992) provide a basis for assessing the effects of lumping discrete reactants. Total surfactant concentrations into and out of a FWS wetland were measured. In addition, liquid chromatography was used to measure alkylbenzenesulfonates of varying alkyl chain lengths, of 11, 12, and 13 carbon atoms. This actual wetland data fits the hypothetical pattern given above, with:

$$k_{11} = 0.59 \text{ per day}, \quad k_{12} = 0.85 \text{ per day}, \quad k_{13} = 1.19 \text{ per day}$$

The hydraulic pattern for the Inaba wetland is not known. Under the presumption that $N = 3$, the effect of the discrete speciation is to reduce the apparent number of TIS to $P = 2.6$. This is less than for the previous hypothetical example, because the spread of the Inaba k-values is less than the spread in the example (c.v. = 0.34 vs. 0.87).

7. Discussion

Historically, the concept of a single contaminant species moving through a wetland at uniform speed has formed the basis of much of the treatment wetland performance analysis. Experimental results have often been interpreted on this presumptive framework, with various models and by various parameter estimation methods. Such data fits are often quite good, in terms of explaining variability in the wetland observations (see for example, Walker, 1995). However, a simplistic level of performance modeling does not typically test the presumptions on which it is based, or evaluate the robustness of the model with respect to wetland characteristics. A data set has been fit, but it is not known whether the model applies to the same wetland at conditions outside the calibration envelope, or to other wetlands of different character. Questions remain concerning transferability of the model, and of the model parameters; and concerning the accuracy of model extrapolation.

The transferability issue may be partially resolved by examining multiple calibrations from a set of wetlands. This procedure has been followed by Kadlec and Knight (1996), who examined and calibrated the PF k–C^* model to multiple data sets. The PF analysis was justified on the presumption that it would yield conservatively low k-values. From that analysis, these authors determined central tendencies for model parameters, and presented frequency distributions of numbers of wetlands across k-values. The breadth of those distributions was unsatisfactorily large, and no intrinsic factors affecting that variability were determined.

Extrapolation of the PF k–C^* model to conditions outside the calibration envelope has been shown to be extremely questionable (Kadlec, 1999). That analysis showed that a de facto TIS wetland would produce data that could be fit extremely well by a PF k–C^* model. However, the resultant parameters were shown to be very sensitive to operating conditions, and very misleading in design calculations. That analysis also showed that PF k-values are not necessarily conservative.

The concepts explored in this paper stem from real and well understood aspects of treatment wetlands. The existence of the distributions examined here is not at issue, but their impact on performance modeling has not been properly acknowledged in the treatment wetland design literature. To differing degrees, the influences of distributed parameters have been set forth in the literature. But despite the knowledge availability, little or no use has been made of it. Best known are the impacts of DTDs on performance modeling, yet in 2001, treatment wetland manuals were still being published with only PFR hydraulic wetland models (Water Environment Federation, 2001).

7.1. Time dependent rate coefficients

The potential effects of speciation in lumped contaminant measures, as manifested in changing rates, have been known for several years (Tchobanoglous, 1969; Crites and Tchobanoglous, 1998). Crites and Tchobanoglous (1998) (C&T) set forth a formulation for a 'retarded rate expression.' This expression is:

$$k = \frac{k_0}{(1 + rt)^n} \tag{32}$$

where, k is reaction rate constant; k_0 is reaction rate constant at time 0 (inlet); n is exponent of retardation; r is coefficient of retardation; t is time (PF travel time), $= x/u$; u is linear water velocity; x is distance along flow direction.

In a broad sense, this could apply for any intrinsic reaction order—first, second, etc. However, the first order presumption is almost always made, and C&T develop their wetland illustration (Example 9–2) for this important case. Further, C&T note that the value of 'n' is approximately 1.0, at least for BOD and TSS. This formulation is to be used in the 'ideal' PF reactor context (see C&T, pp. 133–134).

Here it is shown that this formulation is nothing more than a TIS-type model. The ideal PF model is:

$$u\frac{\mathrm{d}C}{\mathrm{d}x} = -kC = -\frac{k_0}{(1 + rt)^n}C \tag{33}$$

or in terms of time:

$$\frac{\mathrm{d}C}{\mathrm{d}t} = -\frac{k_0}{(1 + rt)}C \tag{34}$$

Integration of this equation yields:

$$\frac{C}{C_\mathrm{i}} = (1 + rt)^{-(k_0/r)} \tag{35}$$

For comparison, the formula for N tanks in series is given by Eq. (15). By comparison of Eqs. (15) and (35), it is seen that they are identical if:

$$r = \frac{k}{N} \tag{36a}$$

$$k_0 = rN \tag{36b}$$

or

$$N = \frac{k_0}{r} \tag{37a}$$

$$k = k_0 \tag{37b}$$

C&T suggest, for BOD, that $r \approx 0.2$ per day, and that $k_0 \approx 0.7–1.0$ per day. Therefore, the equivalent number of TIS is $N = 3.5–5.0$. However, their illustration of the model (their example 9.2) effectively substitutes Eq. (25) of this paper into Eq. (15) of this paper, which is incorrect, leading to an error of 120%.

Shepherd et al. (2001) calibrated a time dependent rate model to COD reduction, with the presumptions of $n = 1$ and PFR hydraulics. As seen above, the PF assumption has a large effect on model parameterization, but Grismer et al. (2001) demonstrated this to be a reasonable assumption for the Shepherd et al. pilot-scale, gravel-filled mesocosms. Therefore, Shepherd et al. (2001), in reality, executed a relaxed TIS model. The values of the TIS parameter computed from their results are $N = 3.4 \pm 1.0$.

7.2. Applicability

The weathering effect associated with speciation produces modeling influences even under uniform conditions in batch processes. Shifting rate constant distributions produce changing mean rate constants, always toward lower values. This inherent slowdown creates modeling artifacts, including false background concentrations and non-exponential decreases in concentration for the mixture. Concentration profiles may be fit very well by a relaxed TIS model, but the parameters involve both hydraulic and speciation effects. Species distributions cause a shift in the apparent number of TIS to lower numbers, to unity and lower.

Nearly all municipal wastewater quality parameters are speciated, and contain combinations of discrete and continuous distributions. TSS represents a lumped category that presumptively has a distribution of settling rates. BOD has particulate and soluble forms, both of which are lumped mixtures. TN has discrete components (ammonium, nitrate), as well as the lumped categories of particulate N and dissolved organic N. TP is also comprised of lumped categories, including particulate, soluble reactive and dissolved organic fractions. It is also likely that fecal coliforms, a lumped collection of microorganisms, exhibit a spectrum of die-off rates.

An important feature of the wetland distribution functions is the width of the distribution, or its variance. The spread determines the magnitude of the effect on a presumed model. The variances (σ^2) of the distributions considered here are:

$$\text{Discrete} \quad \sigma^2 = \sum (k - \bar{k})^2 \tag{38}$$

$$\text{Uniform} \quad \sigma^2 = \frac{(b-a)^2}{12} \tag{39}$$

$$\text{Gamma} \quad \sigma^2 = \frac{1}{n} \quad \text{or} \quad \frac{1}{N} \tag{40}$$

As the DTD becomes narrow, it approaches PF. As the kVDs narrow, the mixture converges to act as single compound. As the model parameters n or N increase, the variance decreases, and the apparent value P approaches N or n, respectively (Table 2). And, as the model parameters n or N increase, the apparent C^* decreases to 0 (Table 3).

It is important to distinguish between stochastic variation and the distributions under consideration here. The distributions considered in this paper are all deterministic. There are also probabilistic components of wetland behavior, which tend to confuse instantaneous observations of performance. Stochastic variability requires some form of filtering before distribution effects can be sorted out (Kadlec, 1997a,b).

8. Conclusions

Numerous treatment wetland studies, using tracer testing, have, without exception, shown that inert materials pass through a wetland with a distribution of speeds. DTDs for wetlands are often responsible, in and of themselves, for non-negligible departures from PF behavior. Those departures in turn create pollutant concentration behavior that cannot be adequately described by the exponential models frequently seen in the treatment wetland literature (e.g. Reed et al., 1995; USEPA, 1999; Water Environment Federation, 2001). Unless strong measures are taken in design to ensure longitudinal compartmentalization or some other form of water redistribution, there will be preferential paths of fast water movement, and the attendant effects on concentrations of contaminants.

Weathering of mixtures produces lower rate constants as treatment proceeds. This effect also invalidates exponential models. The existence of distributions of rate constants produces concentration profiles that are well described by an Eq. (24) that is of the same form as that arising from a TIS DTD (Eq. (15)). It is therefore not necessary to fully understand which of the factors dominates, since the relaxed TIS model can fulfill a role of empirical data fitting. Since it encompasses the end result concentration profiles from a variety of causative distributions, it is better able to represent multiple situations.

The time dependent k-value approach is similar to, but not identical with a gamma distribution of k-values, except for the case of the exponential distribution of rate constants ($n = 1$). However, the concept of clock time as a determinant of a mean rate constant is not intuitive. It seems clear that changing speciation is the cause of time varying rate constant during treatment. Given that premise, then the broader formulation presented here is preferred, because it allows for any type of species distribution coupled with any specified DTD.

Tracer tests form the proper basis for interpreting the disappearance of single compounds. However, tracer testing may not provide the correct parameters for fitting the relaxed TIS model to lumped mixtures.

The examples explored in this paper suggest that either a k–C^* model or a relaxed TIS model will give excellent data fits, and that is indeed the experience of the technology. However, the C^* value in the former reflects several different causes.

There may be a real irreducible component of the substance, or there may be wetland ecosystem feedback of that constituent. But in addition, DTDs and kVDs may create an apparent C^* as an artifact of model parameter fitting. These may be considered 'bypassing C^*' and 'weathering C^*', respectively.

The distribution effects examined in this paper offer a partial explanation of the hazards of either extrapolating beyond the calibration envelope or transferring a model to a second type of ecosystem. As model computations are extended to loadings below those in its calibration, or equivalently to detention times longer than those of the calibration data, there is a possibility of large potential errors. The model may attribute a long-detention plateau to distribution effects, when in fact it is due to a real wetland background that cannot be reduced. Or, a near-plug-flow model calibrated to data from one wetland may not be applicable to another wetland with a different vegetation density distribution.

Distributions of wetland parameters clearly can create modeling anomalies that obfuscate the interpretation and utilization of treatment wetland data. The complexity of wetland processes, some aspects of which have been identified herein, has created two distinctly different reactions among practitioners. Wetland scientists plead for greater quantification and utilization of details (Reddy and D'Angelo, 1997; Wetzel, 2001), but offer no mechanisms for incorporating that detail into practical wetland design. On the other hand, practitioners and regulators decry the 'green box' approach, but reject detailed modeling approaches in favor of rough empiricism (USEPA, 2000). DTD effects are now reasonably well understood, and there is an increasing awareness of the importance of mixture weathering. It is within our reach to sort out and quantify depth and vegetation density.

In the mean time, it seems clear that the PF model should be abandoned unless there is site specific data to defend it. The relaxed TIS model offers the ability to embody both DTD and kVD effects, as well as to encompass the PF extreme by utilization of a large number of apparent TIS.

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