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CHAPTER 6
SUMMARY OF STUDIES ON
MODELING PERSISTENCE OF
DOMESTIC WASTEWATER CHLORINE
IN COLORADO FRONT RANGE RIVERS

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INTRODUCTION

The use of chlorine for partial disinfection of domestic wastewaters typically results in a wastewater treatment plant (WWTP) effluent with several tenths of a mg/L residual chlorine. Since chlorine has been found to be chronically toxic to fish and other aquatic life at a few $\mu\text{g/L}$, that is, a factor of 100 less than typical effluent concentrations, there is a potential for appreciable toxicity to aquatic life from domestic WWTP effluents that are not dechlorinated. The authors have been working with several Colorado Front Range communities in the evaluation of the potential benefits that can be achieved for the receiving water, warm-water sports fisheries, by dechlorinating domestic WWTP effluents.

A hazard assessment approach has been used in these studies. This approach directs the collection of pertinent aquatic toxicology and chemistry information necessary to define the hazard that a particular contaminant discharge represents

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to the beneficial uses of the particular water under investigation. A key component of hazard assessment studies is the assessment of the fate-persistence of the contaminant in the receiving waters under various hydrologic, climatic, and other conditions that exist at that location.

As part of these hazard assessment studies, an aquatic chemistry modeling approach has been developed to define the concentrations of residual chlorine discharged by several Colorado Front Range WWTPs in the respective receiving waters, as a function of distance downstream from the points of discharge. This paper presents a summary of these studies. A more detailed discussion of the experimental methods, modeling approach, and results can be found in Heinemann¹ as well as in other works of Lee and his associates.²⁻⁶ In addition, a summary of the hazard assessment studies has been presented by Newbry et al.⁷

MODELING APPROACH

Equation 1 describes the major factors that impact chlorine dissipation in a river and that were considered in this modeling effort:

$$\begin{aligned} (dC/dt)_{\text{overall}} = & (dC/dt)_{\text{dilution}} + (dC/dt)_{\text{demand}} + \\ & (dC/dt)_{\text{phototransformation}} + (dC/dt)_{\text{volatilization}} \end{aligned} \quad (1)$$

where C = chlorine concentration
t = time

A series of studies was conducted in four of the six Colorado Front Range receiving waters below domestic WWTPs investigated to determine the reaction orders and rate constants for these factors. Several "time-lag" tests were run in which subsamples of river water samples collected near the WWTP outfalls were analyzed for chlorine periodically over a several-hour period. In samples maintained in darkness and at constant temperature, concentrations consistently decreased according to first-order kinetics, indicating that the exertion of chlorine demand was a first-order process. Chlorine concentrations in similar subsamples exposed to sunlight decreased at consistently faster rates than in those kept in the dark, but they also decreased according to first-order kinetics. This indicated that phototransformation of chlorine was a first-order reaction, a finding consistent with what is reported in the literature.¹

First-order kinetics is usually assumed for volatilization in most environmental fate models in which this factor is included. Nothing was found at any of the six

study sites that would contradict the applicability of this assumption for the Colorado Front Range rivers.

The assumption that dilution is a first-order process may not always be appropriate, but as long as the ultimate dilution factor is known, inaccuracies caused by this assumption will only appear in the dissipation estimates for the zone of physical mixing. However, in this study the first-order dilution rate assumption provided acceptable predictions of chlorine concentrations in the zone of mixing.

Since it was believed that each of the model components could be suitably described by first-order kinetics, it was assumed that the overall dissipation of chlorine in the rivers studied could be described by first-order kinetics. The comparison of model output with measured chlorine concentrations supported the validity of that assumption.

Dilution

The rate constants for dilution (k_{dil}) were estimated using the mean effluent chlorine concentration, the concentration calculated for the point at which the particular effluent and river water were completely mixed, the mean velocity of the river determined using drouges-tracers, and the length of the zone of mixing of the effluent with the river. The dilution rate constants were calculated according to the equation

$$C_z = C_o e^{-k_{dil} t_z} \tag{2}$$

where C_z = chlorine concentration at end of zone of mixing (see Equation 3)
 C_o = chlorine concentration in WWTP discharge
 t_z = travel time to end of zone of mixing (length of zone of mixing ÷ river velocity)

The term C_z is calculated as

$$C_z = \frac{(C_o)(Q_o) + (C_u)(Q_u)}{(Q_o + Q_u + Q_m)} \tag{3}$$

where in addition to the terms defined above,

- Q_o = WWTP flow
- C_u = chlorine concentration above discharge point
- Q_u = flow in river above discharge point
- Q_m = other water input flows within zone of mixing

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$$K_2 = 2.98 (U/H^{1.5})^{0.703} \text{ (in days}^{-1}\text{)} \text{ (Padden and Gloyna}^{11}\text{)} \quad (4)$$

The chlorine demand and phototransformation rate constants were estimated using controlled condition "bottle tests." For each test set, BOD bottles were filled with river water containing WWTP effluent collected as a single sample. Half of the bottles had been blackened with electrical tape to eliminate light; half were left transparent. Three of each type of bottle were suspended in the river water from a six-arm support (trapeze) which kept the bottles separated. The only cause for a decrease in chlorine concentration in the dark bottles was the exertion of demand by certain components of the WWTP effluent and river water (such as dissolved or particular organics), whereas the chlorine in the light bottles could undergo phototransformation as well as exertion of chemical demand. One of each type of bottle was removed periodically over a several-hour period for chlorine determination. The rate constant for chlorine demand was determined by plotting on semi-log paper the chlorine concentration in the dark bottles as a function of time. The demand rate constant, k_{dem} , was the slope of the line of best fit. Similarly, the difference in chlorine concentration between the light and dark bottles was plotted as a function of time to obtain the phototransformation rate constant, k_{phot} . Chlorine demand in the bottles was assumed to be the same as that in the river, since the bottles were maintained at the temperature of the river water, and the incubation periods were kept short. Phototransformation in the light bottles was assumed to be the same as that in the river, since the bottles were shaded by the light attenuation properties of the water, and the placement of the bottles was such that they were located near the position of geometric mean light intensity in the water column. The assumption that the light attenuation by the BOD bottles themselves is negligible is frequently made in the water quality field in the evaluation of photosynthesis by the light- and dark-bottle technique.

Volatilization

Volatilization rate constants were estimated by the method of Tsivoglou,⁸ who showed that the rate of volatilization of a compound can be estimated by the oxygen transfer rate (K_2) of the water body and the relative molecular diameters of the compound and oxygen. The oxygen transfer rate (base e) was estimated using the average-depth, average-velocity curves presented by Zison et al.⁹ The chlorine volatilization rate constant ($k_{2,Cl}$) was calculated using the following equations,

$$K_2 = 0.344 (U^{0.67}) (H^{-1.85}) \quad (\text{Owens et al.}^{12}) \quad (5)$$

$$k_2 = 2.303 K_2 \quad (6)$$

$$k_2' = k_2 \theta^{(T_2 - T_1)} \quad (\text{Metcalf and Eddy, Inc.}^{10}) \quad (7)$$

$$k_{2,Cl} = k_2' (1.06) (d_{O_2}/d_{Cl})^{1.26} \quad (\text{Tsvoglou}^8) \quad (8)$$

where in addition to the terms defined above,

- U = mean velocity (ft/s)
- H = mean depth (ft)
- K_2 = oxygen transfer rate constant (base 10 at 20°C)
- k_2 = oxygen transfer rate constant (base e at 20°C)
- k_2' = oxygen transfer rate constant (base e at ambient temperature, °C)
- $k_{2,Cl}$ = chlorine volatilization rate constant (base e at ambient temperature, °C)
- θ = volatilization rate constant temperature correction factor = 1.024
(Metcalf and Eddy, Inc.¹⁰)
- T_1 = 293 K
- T_2 = ambient temperature, K
- d_{O_2} = molecular diameter of $O_2 = 2.82 \text{ \AA}$
- d_{Cl} = molecular diameter of chloramine = 5.4 \AA

Equation 4 was used for the studies on the Arkansas River; Equation 5 was used for all other studies. The difference is due to the difference between the depth of the Arkansas River system and the other systems.

In some instances the volatilization rate was checked by computing the difference between the decrease in chlorine concentrations in the light-bottle tests and the die-away found in the river, since volatilization was the only primary means of dissipation not accounted for by dilution or in the bottle tests. The overall chlorine dissipation rate constant (k_R) was calculated according to the equation

$$C_2 = C_0 e^{-(k_{dil} t_z) - (k_R t_z)} \quad (9)$$

where in addition to the terms defined above,

- C_2 = chlorine concentration at end of river reach
- t_z = time of travel to end of river reach

The light-bottle dissipation rate constant was then subtracted from the k_R value to obtain the volatilization rate estimate.

RESULTS

Complete data from the chlorine persistence studies have been presented by the authors elsewhere^{2-4,6}. The first-order rate constants for each of the dissipation mechanisms determined for the various study locations are presented in Table I as averages for the study reaches. When necessary, the river reach investigated was divided into sections based on physical characteristics, and individual rate constants for each reach were computed; the individual rate constants, rather than the overall constant reported, were used in model predictions. There was little obvious pattern in the rate constants with station or season.

Table I. Average Rate Constants Determined for Chlorine Dissipation

Location	Rate Constants, base e (min ⁻¹ @ 20°C)			
	k _{demand}	k _{phototransformation}	k _{volatilization}	k _{dilution}
Fort Collins				
WWTP No. 1				
Winter	0.0069	0.0008 (est)	0.0065	0.0246
Spring	0.0068	0.0025	0.0059	0.0135
Summer	0.0046	0.0046	0.0389	0.0200
Fort Collins				
WWTP No. 2-River				
Fall	0.0165	0.0100 (est)	0.0076	0.3232
Winter	0.0165	0.0020	0.0117	0.1911
Spring	0.0068	0.0025	0.0036	0.5159
Fort Collins				
WWTP No. 2-Ditch				
Fall	0.0165	0.0100 (est)	0.0870	N/A
Winter	0.0069	0.0020	0.0294	0.7519
Summer	0.0036	0.0022	0.0061	0.0013
Loveland WWTP				
Winter	0.0114	0.0020 (est)	0.0106	0.1047
Spring	0.0114	0.0026	0.0139	0.1309
Summer	0.0124	0.0147	0.0101	0.3323
Pueblo WWTP				
March	0.0012	0.0004	0.0019	0.0172
Colorado Springs				
September (est)	0.0012	0.0004	0.0038	
Winter (@ 5°C)	0.0008	0.0004	0.0027	

Chemical demand was responsible for 10-50% of the total dissipation of chlorine; phototransformation, from 5-30%; and volatilization, from 20-80%. The percentage ranges were all about the same. The overall reactivity rate constants found in these studies were not unlike those reported in the literature.^{1,2} The rate constants presented in Table I, however, have all been computed for a river temperature of 20°C (except for phototransformation, for which the rate is generally considered to be independent of temperature). The actual rate constants used for various times of the year will be different from those presented according to the water temperatures encountered. Thus the summer rate constants in the Colorado Front Range systems would be about the same as those presented or higher; whereas winter rates, due to lower temperatures, would be lower than those presented. Rate constants can be converted from one temperature to another through the van't Hoff-Arrhenius equation given below:

$$K_2 = K_1 \theta^{(T_2 - T_1)} \quad (10)$$

where K_2 = rate constant at temperature T_2
 K_1 = rate constant at temperature T_1
 T_2 = temperature desired (K)
 T_1 = temperature given (K)
 θ = temperature correction coefficient

The value for θ for volatilization (1.024) was obtained from Metcalf and Eddy, Inc.¹⁰ The values for θ for demand temperature correction were determined by measuring the demand rate constants at different temperatures. A value of 1.099 was used in the fall, winter, and spring studies, and a value of 1.035 was determined for the summer studies.

A word of caution should be given on the use of these rate constants for other systems. As shown in Table I, there are marked changes in the constants from season to season; factors such as stream discharge and velocity, water depth, amount of shading of the stream, algae and periphyton, etc., affect the rate constant values. It should also be noted that the mixture of residual chlorine compounds present in the study site waters was typical of those compounds found below the discharge of chlorinated, nonnitrified, domestic WWTP effluents. It is likely that somewhat different chlorine residual persistence would be obtained for residuals derived from chlorination of a highly nitrified effluent, which would have a substantial amount of the chlorine residual in the form of organic chloramines.

Because of the frequently encountered variability in chlorine concentrations in the WWTP effluents studied, it was necessary to normalize the effluent chlorine concentrations to 1.0 for modeling and data manipulation. In general, the

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concentrations of chlorine in a water mass were measured at each study location as the water mass moved downstream from the discharge. The data were normalized by dividing downstream concentrations averaged at each station by the average effluent concentration (yielding a normalized effluent concentration of 1.0). Figures 1-4 are representative of the model predictions and corresponding normalized chlorine concentrations found with distance (expressed as travel time) downstream of the WWTP effluent discharges evaluated. The normalized, measured concentrations with their 95% confidence limits are shown as the solid or open circles and associated confidence interval bars; the predicted concentrations based on dilution alone, as well as on both dilution and reactivity, are shown as the broken and solid lines respectively. As expected, these figures show that dilution is the dominant factor causing chlorine concentration reductions

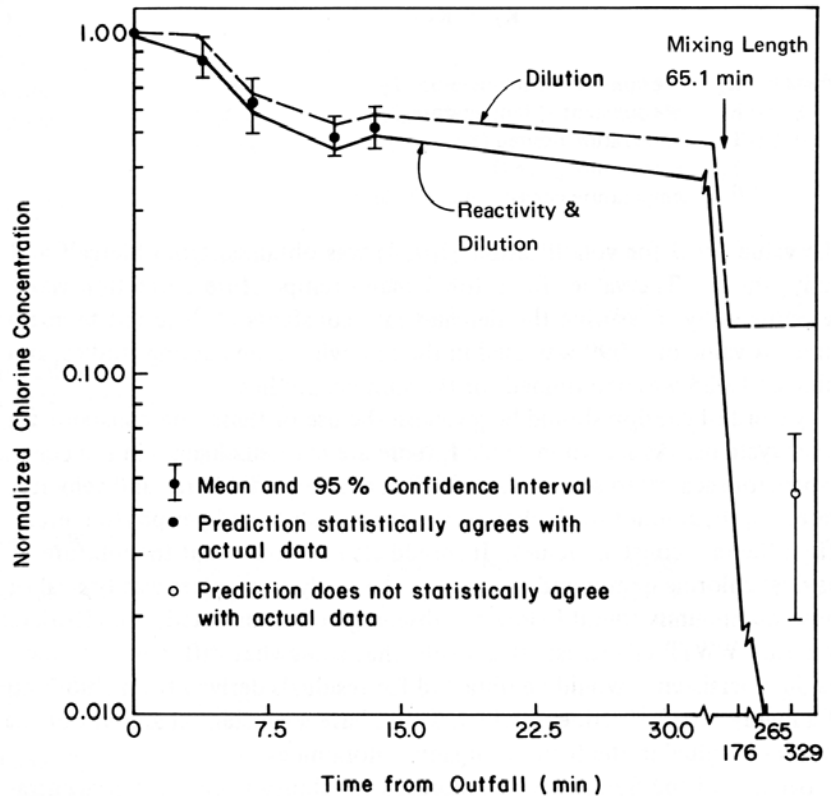


Figure 1. Normalized measured chlorine concentrations and model predictions at Fort Collins WWTP No. 1, Poudre River (spring 1980).

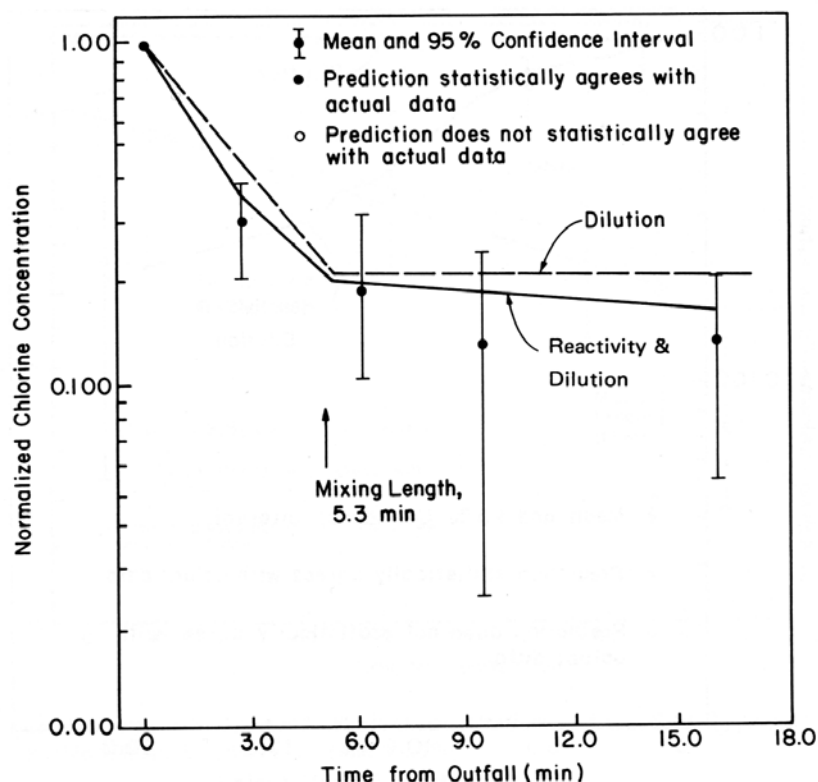


Figure 2. Normalized measured chlorine concentrations and model predictions at Fort Collins WWTP No. 2, Poudre River (fall 1979).

in the zones of physical mixing between the effluent and river water, whereas the reactivity is responsible for the ultimate dissipation of chlorine to non-detectable levels beyond this zone. The model prediction lines for most of the study areas were jagged, that is, had several changes in slope. This was caused by changes in the physical characteristics of the receiving water with distance downstream, such as rate constants and river depth, which changed the model inputs.

Figure 1 shows the predicted and actual effluent chlorine behavior in the Poudre River downstream of the Fort Collins WWTP No. 1 during spring low-flow conditions. Within the mixing length, the concentration was predicted well by the model; the one sample collected beyond the zone of mixing had a higher concentration than predicted. The investigators believed that the modeling weak-point in this case was the river velocity estimation and/or the effluent chlorine demand rate constant used as model input.

Model predictions for the Poudre River below the Fort Collins WWTP

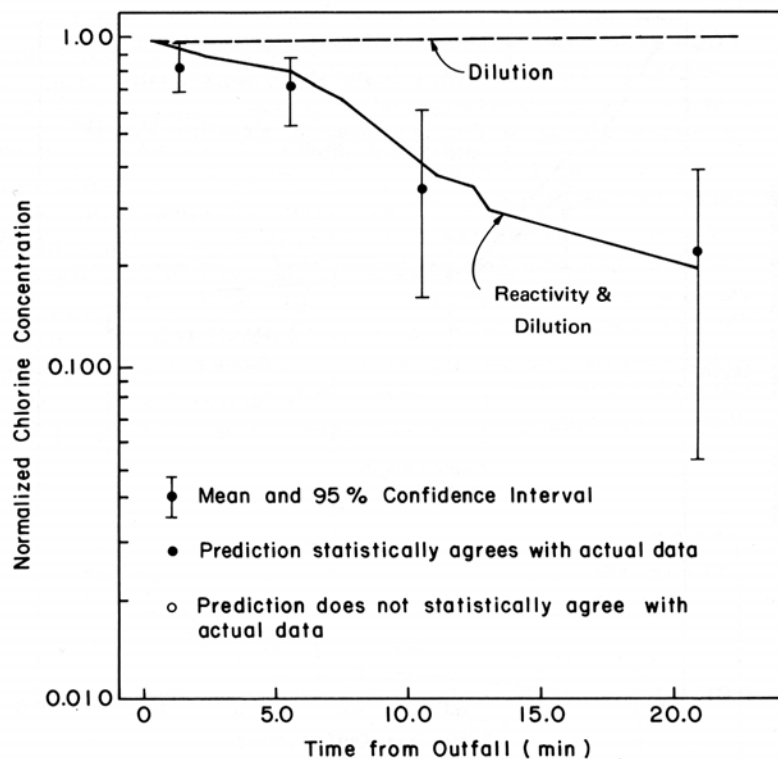


Figure 3. Normalized measured chlorine concentrations and model predictions at Fort Collins WWTP No. 2, Fossil Creek Ditch (autumn 1979).

No. 2 are presented in Figure 2. During all three seasons evaluated in this reach of the river, including the fall conditions illustrated, the chlorine concentrations were predicted quite well by the model, rate constants, and other input parameters used.

Figure 3 shows the model predictions of the behavior of the Fort Collins WWTP No. 2 effluent chlorine in Fossil Creek Ditch. The close agreement between predicted and measured values was attributed to two factors. First, there was essentially no dilution water, which meant that chlorine disappearance was due entirely to chlorine reactivity. This made the modeling simpler and the dissipation predictions straight-line. Second, Fossil Creek Ditch in the study area was straight and fairly uniform in shape, making extensive adjustments for physical irregularities unnecessary. The goodness of fit in this, the simplest of all the study sites, lends credibility to the model itself and points to the role that the physical characteristics of the river reach can play in this modeling.

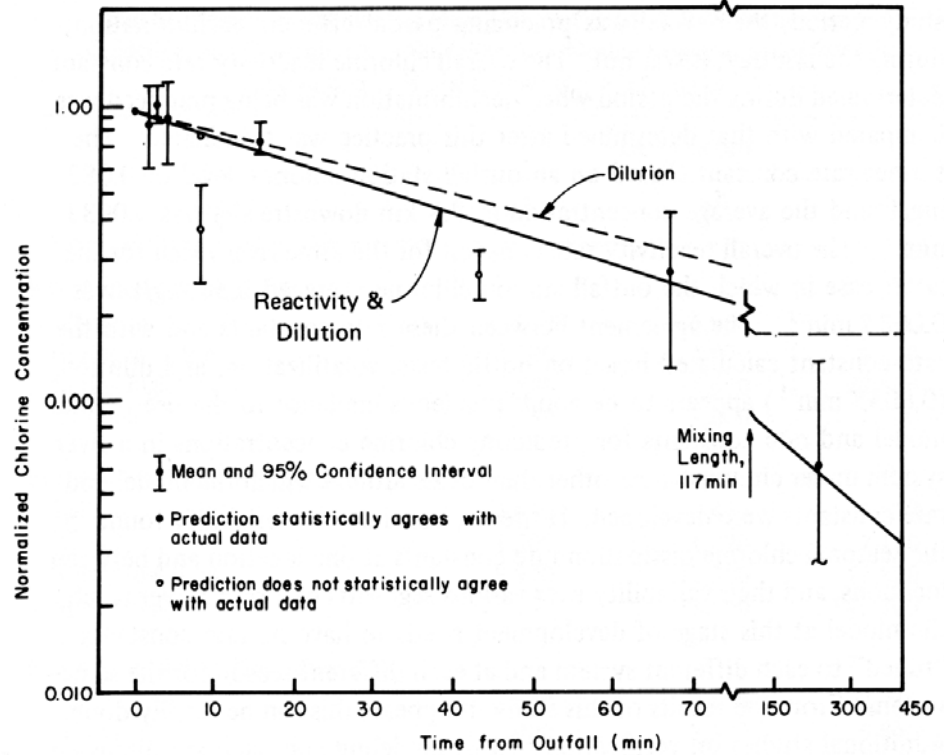


Figure 4. Normalized measured chlorine concentrations and model predictions at Pueblo WWTP, Arkansas River (March 1980).

Figure 4 shows model chlorine predictions for the Arkansas River down-stream of the Pueblo WWTP within the zone of physical mixing and beyond. It illustrates that the model with rate constants determined as outlined herein provides close estimations of the chlorine concentrations found in the Arkansas River downstream of the Pueblo WWTP discharge.

MODEL VERIFICATION

For any model to be used to make predictions of future conditions or for other systems, it must be verified. The demonstrated applicability of the modeling approach to the various Colorado Front Range rivers provides a degree of model verification in that, by using the same testing procedures for rate constant development, the models tracked the measured chlorine concentrations downstream of most of the effluent discharge points evaluated.

There was an opportunity in the Pueblo WWTP system to verify the model for a single system under altered effluent chlorine concentrations conditions without altering the rate constants. During the first part of the study period, the WWTP was

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practicing partial effluent dechlorination; during the last day, it was not. The overall chlorine reactivity rate constant determined during the period when dechlorination was being practiced was compared with that determined after this practice was terminated. The former rate constant (based on an outfall station chlorine level of 0.082 mg/L and the average concentration at 8.8 km downstream) was 0.0033 min^{-1} . The overall reactivity rate constant for the same river reach for the latter case in which the outfall station chlorine averaged 0.56 mg/L was 0.0035 min^{-1} . The agreement between these rate constants and with the rate constant calculated based on bottle tests, volatilization, and dilution (0.0035 min^{-1}) appears to be good; this lends credence to the use of the model and rate constants for predicting chlorine concentrations in a river system under circumstances other than those around which the model and rate constants were developed. However, based on the variability found in the seasonal chlorine dissipation rate constants at one location and between locations, and their variability over various segments of a single river reach, the model at this stage of development needs to have its rate constants "tuned" to each different system and at each different season for the same system. From the results of this study, it appears this can be readily done. Additional studies on verification of this modeling approach are discussed by the authors.^{1,2,4,6}

USE OF MODEL IN WATER QUALITY EVALUATION

The results of this modeling can play a vital role in the evaluation of the potential impact of the chlorine in a chlorinated WWTP effluent (and in principle, for any other type of discharge) on receiving water quality. This was demonstrated by Lee et al.²⁻⁶ and Newbry et al.,⁷ who used model results to estimate lengths of the river reaches in which the chlorine concentrations exceeded various water quality guideline values for chlorine under given river and effluent conditions. The approach can be used to evaluate the impact of reducing the chlorine concentration of a particular effluent by any given amount on the river distance over which chlorine will persist at any level, for example, how far acutely toxic levels may be found or how far levels above the chronic safe concentration would be found. This model was used by Newbry et al.⁷ for this purpose; Table II was produced from the model output. It illustrates the sizable differences that occur in the sizes of the zones in which any particular guideline level is exceeded, depending on season and flow.

The rate constants for chlorine dissipation have been developed and used herein without regard to certain factors (except temperature) which can alter their

Table II. Distances Required to Meet USEPA Criterion, AFS-Recommended Guideline, and Colorado State Guideline for Chlorine Downstream of WWTPs, According to Model Output ($C_O = 0.5$ mg/L)

Study	Distance (m)		
	A ^a	B ^b	C ^c
Fort Collins WWTP No. 1			
Winter	5,570	6,900	7,880
Spring low flow	3,180	4,070	4,890
Spring high flow	2,880	15,400	27,400
Summer	1,160	1,030	1,700
Fort Collins WWTP No. 2-River			
Fall	2,640	3,410	3,970
Winter	1,500	1,870	2,150
Spring low flow	9,990	13,300	15,800
Spring high flow	20,750	32,500	41,200
Fort Collins WWTP No. 2-Ditch			
Fall	1,680	2,510	2,750
Winter	6,900	8,570	9,840
Summer	5,800	6,990	7,870
Loveland WWTP			
Winter	4,060	4,840	6,350
Spring	2,820	3,730	4,410
Summer	1,350	1,900	2,300
Pueblo WWTP			
	13,213	20,899	26,572
Colorado Springs			
Summer (est)	21,700	26,400	29,400
Winter (est)	30,200	36,700	41,500

^aA = to 0.01 mg Cl/L – USEPA Red Book criterion.

^bB = to 0.005 mg Cl/L – AFS recommended guideline.

^cC = to 0.003 mg Cl/L – Colorado guideline.

effective values and use in field applications. Although these factors, which are discussed below, were not considered in the computations, the predictions made by the model were, in general, good; the real impact of these factors on the rate constants may be minor, or some of them may have been avoided by the sampling pattern.

Phototransformation does not occur at night. Therefore, computations of

chlorine dissipation should be made at night, excluding the phototransformation mechanism. The potential importance of the lack of phototransformation at night can be demonstrated by comparing the extent to which the chlorine from the Colorado Springs WWTP persists at levels greater than 0.003 mg/L in Fountain Creek in the summer if there were phototransformation and if not. Even with the relatively small phototransformation rate constant assumed for Fountain Creek (0.0004 min^{-1}), the lack of phototransformation under the summer conditions would result in an extra kilometer of the river having concentrations of chlorine above 0.003 mg/L (in addition to the 27 km already exceeding that level). Variations in pH are also reported to impact phototransformation rates.¹ With increasing pH, the quantum yield increases, which increases phototransformation; this may be of significance in rivers in which, due to algal growth or other causes, the pH varies over a daily cycle. Differences in phototransformation due to sunlight intensity-angle of the sunlight-will necessitate estimating this rate constant for each season. Deviations caused by cloud cover may also necessitate determination of a range in phototransformation rate constants for any system.

WWTP flows are generally less at night, and the effluent composition is different from that during the day: these are factors which could impact day and night chemical demand differences, as could pH variations, over a diel cycle.

NEED FOR FURTHER WORK

To use this modeling approach at present, light- and dark-bottle experiments must be run in the river for phototransformation determination. Aside from this, all other determinations, including tests for chlorine demand, can be readily made in the laboratory or obtained from existing records. Therefore, there is a need to conduct additional studies to relate phototransformation measurements to actinometer readings and eventually to a simple parameter such as Secchi depth or turbidity in the river. If such correlations can be developed, a measurement of river turbidity or Secchi depth can be mathematically converted to a phototransformation rate constant, eliminating the need for in situ experiments. Further, work needs to be done to evaluate the impacts of normal diel fluctuations of temperature, pH, sunlight (cloud cover), shade, flow, and WWTP characteristics on the persistence of chlorine and on its significance to aquatic life.

SUMMARY AND CONCLUSIONS

A modeling approach has been developed based on several Colorado Front Range streams in the vicinity of chlorinated WWTP effluent discharges to estimate the persistence of chloramines in natural water systems. It was found through a series of simple measurements and tests that rate constants can be developed to describe the dissipation rate of chlorine and its dilution so that estimates can be made of the persistence of the chlorine downstream of the WWTP discharges. This approach reliably predicts chlorine persistence in most sites evaluated and has been verified to a limited extent in Colorado Front Range streams.

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