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Estimating fast and slow reacting component in surface and groundwater using 2R model

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Abstract

Maintaining residual chlorine levels in a water distribution networks is a challenging task; especially in the context of developing countries where water is usually supplied intermittently. To model chlorine decay in water distribution networks, it is very important to understand chlorine kinetics in bulk water. Recent studies suggested that chlorine decay rate depends on initial chlorine levels and type of organic and inorganic matter present in water, indicating that first order decay model is unable to accurately predict chlorine decay in bulk water. In this study, we employed two reactant model (2R) to estimate the fast and slow reacting components in surface water and groundwater.

- We carried out bench scale test for surface and groundwater at initial chlorine level of 1, 2 and 5 mgL⁻¹. We used decay datasets to estimate optimal parameter values for both surface water and groundwater. After calibration, the 2R model was validated with two decay dataset with varying initial chlorine concentration (ICC). This study came up with three important findings (a) the ratio of slow to fast reacting components in
- ¹⁵ groundwater was thirty times greater than that of the surface water, (b) 2R model can accurately predict chlorine decay in surface water, 98 % of the variance in the chlorine decay test was explained by the model and (c) in case groundwater, 2R model prediction accuracy reduced with the decrease in ICC levels, only 87 % variance in data was explained by the model. This could be attributed to high slow to fast reactant ratio in 20 groundwater.

1 Introduction

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The presence of 0.2 mg L⁻¹ of residual chlorine in drinking water in known to reduce public health risks significantly (Arnold and Colford, 2007; Pattanayak et al., 2005). One of the key tasks of water managers worldwide and especially in developing nations is to maintain residual chlorine levels of drinking water in distribution systems. This requires much higher concentration at entry point in order to ensure that the minimal residual





chlorine concentration of 0.2 mg L^{-1} is retained at any point of time before reaching the end consumers.

To maintain 0.2 mg L^{-1} of residual chlorine in water, sodium hypochlorite or liquid chlorine is added to the secondary treated water. Before water reaches the consumers,

- ⁵ part of this chlorine is lost to reactions with the organic and inorganic matter present in water after secondary treatment, as well as to reactions with biofilms and corrosion products present within the distribution network (Al-Jasser, 2007; Hallam et al., 2002; Helbling and VanBriesen, 2007). Any excessive addition of chlorine to water leads to harmful by-products which pose risks to public health, therefore it becomes very im-
- ¹⁰ portant for water managers to optimize the dose of chlorine added to water ensuring that the right levels of residual chlorine is retained in the distribution networks (Hrudey, 2009; Richardson, 2003; Singer, 1999).

First-order decay process is generally employed to simulate chlorine decay within the distribution network (Hua et al., 1999; Vasconcelos et al., 1997; Rossman et al.,

- 15 1994). The model assumes that chlorine decay rate is a function of chlorine levels within the bulk water. However recent studies have shown that in addition to chlorine levels other factors like the type of organic/inorganic matter present, temperature, pipe material also affect chlorine decay rates in the distribution system (Al-Jasser, 2007; Hallam et al., 2002; Mutoti et al., 2007).
- The total chlorine decay within the water distribution network is caused by (a) chlorine reaction in bulk water and (b) with the biofilm attached to the pipe surface also termed as wall decay (Hallam et al., 2002; Powell et al., 2000 Al-Jasser, 2007). For prediction of chlorine decay over time, models require accurate estimation of chlorine reaction in bulk water and with the biofilm on pipe wall surface. Pilot loop setups/simulators
- are employed to estimate the contribution of wall reaction to chlorine decay (Frias et al., 2001; Lehtola et al., 2006; Rossman, 2006; Rossman et al., 1994). This is achieved by subtraction of bulk reaction rate from the total chlorine decay in the pipe loop. Various authors have argued about the need to accurately model bulk decay before making an attempt to estimate the contribution of wall decay (Fisher et al., 2011).





Chlorine kinetics depends on the type and amount of dissolved organic matter (DOM) and inorganic matter present in the bulk water. DOM in water is derived primarily from decaying organisms such as plants or algae, and is often classified into humic and non-humic substances such as proteins, carbohydrates and lipids. Generally speaking,

- ⁵ DOM from marine and aquatic sources is more enriched in aliphatic structures while DOM from terrestrial/higher plant sources is rich in aromatic compounds (Chen et al., 2010). The inorganic compounds in surface water are derived from the dissolution of minerals present in the bed rock as water flows, whereas groundwater dissolves minerals as it percolates through the vadose zone as well as during its stay in the saturated zone. Therefore in comparison to surface water, groundwater contains high
- 10 saturated zone. Therefore in comparison to surface water, groundwater contains high levels of in-organic substances such and nitrates, manganese, arsenic and iron.

Several studies have shown that in homogeneous systems chlorine exhibits faster reaction rates with ammonia, sulphates, nitrates, nitrites, arsenic and iron as compared to Manganese (Mn (II)) (Deborde and Von Gunten, 2008). Therefore given different

characteristics of components (organic and inorganic) present in surface and groundwater, the precursors to chlorine reactions can be divided into fast and slow reacting fractions (Gallard and von Gunten, 2002). Table 1 presents the list of possible fast and slow reacting components in different types of water.

The two reactant model is a simplified second-order decay model which uses notional fast and slow reacting agents involved in second order reactions with chlorine over long travel periods within the distribution network (Fisher et al., 2011). The second order reaction rates and the resulting 2R model is given by the following equation

$$\frac{dC_{f}}{dt} = -K_{f} \cdot C_{f} \cdot C_{cl}$$

$$\frac{dC_{s}}{dt} = -K_{s} \cdot C_{s} \cdot C_{cl}$$

$$\frac{dC_{cl}}{dt} = \frac{dC_{f}}{dt} + \frac{dC_{s}}{dt}$$
(3)

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where, C_{cl} is the concentration of residual chlorine levels (mgL⁻¹), C_{f} and C_{s} are the concentrations of fast and slow reducing agents (mgL⁻¹) and K_{f} and K_{s} are the fast and slow reaction rate coefficients (Lmg⁻¹ h⁻¹).

The initial concentration of fast and slow reactant (C_{os} and C_{of}) and their respective decay coefficients (K_{f} and K_{s}) can be estimated straight forwardly using AQUASIM software (Fisher et al., 2011).

So far 2R model has been calibrated and tested for surface water under varying conditions such as temperature variation, type of treatment and re-chlorination (Fisher et al., 2011; Mutoti et al., 2007; Rossman, 2006). In this study, we employed 2R model

- to estimate optimal parameters for prediction of residual chlorine in both surface water and groundwater. The model was calibrated separately for test water with initial chlorine levels ranging from 1 to 5 mg L⁻¹. To establish the suitability of 2R model, we validated it against two chlorine decay datasets. One decay dataset was the subset of the calibration dataset and the other dataset was purposely reserved for validation.
- ¹⁵ This study also allowed us to check the feasibility of 2R model in predicting chlorine decay for different types of water.

2 Methodology

We first calibrated 2R model against whole dataset for two types of water: surface water and groundwater to obtain a single invariant set of four parameters that characterize water. The model was then validated by comparing the model estimates with the decay test data. The characteristics of experimental data set are presented in Table 2.

2.1 Chlorine decay test data

Chlorine decay test data for calibration and validation of 2R model was obtained by conducting bench scale residual chlorine decay test at the ATREE Water and Soil Lab-

²⁵ oratory.

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Groundwater and surface water samples were obtained from respective sources and water quality characteristics were determined as presented in Table 3. The test water was stored in amber glass bottles while sodium hypochlorite was added to attain desired initial chlorine concentrations (ICCs). The bench scale tests for both types of water were run at ICC – 1, 2, 4.5 and 5 mgL⁻¹. The amber glass bottles were kept in an incubator set at ambient temperature (25 to 30° centigrade). Hourly samples were drawn from the bottles and free chlorine levels were measured for 72 h. Free chlorine in the test water was measured using Merck Spectroquant[®] Picco colorimeter. The free chlorine measurement range of the instrument is 0.01 to 6.00 mgL⁻¹ (APHA, 2005).

¹⁰ The decay test results obtained were within the chlorine measurement range of the instrument.

2.2 Model parameter estimation

AQUASIM package was used to estimate optimal parameter values for 2R model to obtain the best fit for the three decay dataset (ICC – 1, 2 and 5 mgL⁻¹; bench scale laboratory experiments). Optimal parameters are the values that allow the model to predict chlorine decay in each type of water accurately. As explained by Fisher et al. (2011) AQUASIM software calculates the sum of squared differences between the each experimental data point and the corresponding model prediction, assuming an initial set of parameter values. This sum is derived by the variance of the data to form a chi – square (χ^2). Using the simplex technique it then systematically varies the parameter values to search for a set that produces the best fit (minimizes χ^2).The coefficient of determination (R^2) was also obtained which would indicate the total variance in dataset explained by 2R model regardless of the distance between the decay curves.

We calibrated 2R model using chlorine decay test data for initial chlorine levels of 1,

²⁵ 2 and 5 mg L⁻¹. The optimal parameters for groundwater and surface water were determined separately. After calibration, the 2R model was validated using two datasets i.e. of ICC $- 2 \text{ mg L}^{-1}$ (a subset of calibration dataset) and ICC $- 4.5 \text{ mg L}^{-1}$ (independent chlorine decay dataset).





3 Results

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3.1 Chlorine decay and water quality

The results of bench scale chlorine decay test are presented in Fig. 1. The graph presents the fraction of chlorine remaining in test water at different ICCs. The data presents two interesting findings: (i) the decay rate decreases with increase in initial chlorine levels and (ii) the chlorine decay rate in surface water is greater than that of groundwater.

The above results suggest that first-order decay model is unable to predict chlorine decay in bulk water and the chlorine decay rate depends on the ICC and level of or-¹⁰ ganic/inorganic matters present in test water (Fisher et al., 2011). So far 2R model have successfully been employed to predict chlorine decay in surface water. In the next section, we employ 2R model to estimate fast and slow reacting components is test waters. In addition to this we will also test the feasibility of 2R model in predicting chlorine decay in groundwater. The chlorine decay data presented in Fig. 1 is used for calibration of 2R model using AQUSIM software.

3.2 2R model calibration and validation

We calibrated and validated 2R model by using decay data from bench scale laboratory studies. The 2R model for groundwater and surface water was calibrated separately using the dataset for ICC – 1, 2 and 5 mg L^{-1} . Figure 2 presents the calibrated dataset for groundwater and surface water.

2R model predicted optimal values for four parameters simultaneously by minimizing chi-squared (χ^2) value. Minimization of χ^2 was readily achieved using optimization technique available within AQUASIM parameter estimation procedure. The optimal values derived are presented in Table 4 with associated χ^2 value.

²⁵ Figure 2 presents the decay curves obtained from the simulation using optimized parameters values for the calibration dataset. We observed good agreement between





chlorine residual data and the model estimates for all ICCs, both for surface water and groundwater.

For surface water, 2R model underestimated chlorine levels for 0 < t < 60 h for decay sets ICC -5 mgL^{-1} and ICC -1 mgL^{-1} whereas for ICC -2 mgL^{-1} the chlorine levels were underestimated for 0 < t < 20 h. In case of groundwater, chlorine decay was underestimated for ICC -5 mgL^{-1} and ICC -2 mgL^{-1} for 0 < t < 60 h and for ICC -1 mgL^{-1} chlorine was underestimated for 0 < t < 5 h. Although the χ^2 values for surface water and groundwater was 0.18 and 0.48 respectively, the R^2 values were greater than 0.98 for surface water and 0.99 for groundwater indicating that only 2 and 1 % variance in the calibration dataset was unexplained by the model. We also checked the validity of our results by comparing optimal parameter values obtained from our dataset with that of the other studies. Table 5 presents the optimal parameters obtained for other

test waters. The χ^2 values obtained for our dataset was comparable to that of the experimental dataset from other studies (Fisher et al., 2011).

- We observed that in groundwater the ratio of slow to fast reacting component is thirty times greater than that for the surface water. This suggests that as groundwater travel through vadose zone most of the fast reacting organic matters is consumed by the microorganism leaving behind the non-biodegradable organic/inorganic matters (slow reacting component) (He et al., 2006; McCarty et al., 1981). The slow reacting components are difficult to oxidize therefore high levels of chlorine residual over time
- ²⁰ components are difficult to oxidize therefore high levels of chlorine residual over time were observed in groundwater.

3.3 Validation of 2R model

The 2R model was fitted for experimental dataset (a) surface water (ICC - 2.6 and 4.2 mgL⁻¹), (b) groundwater (ICC - 2 and 4.5 mgL⁻¹) as shown in Fig. 3.

First we calibrated 2R model with decay test commencing highest and lowest ICC, as the extrapolation outside the calibration range is less reliable. Then we validated the model by using two chlorine decay datasets i.e. one from the calibration dataset (ICC





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 -2 mg L^{-1}) and other dataset that has not been used for the model calibration (ICC -4 mg L^{-1}).

The R² value obtained for both the datasets of surface water was greater than 0.98 indicating that only 2% of the variance is unexplained by the 2R model. This suggests
the suitability of 2R model for chlorine prediction in surface water. In case of groundwater, R² value 0.89 and 0.94 was obtained for decay test at ICC – 2 mgL⁻¹ and ICC – 4.5 mgL⁻¹ respectively. The accuracy in chlorine prediction increased with increase in ICC which could be attributed to high slow to fast component ratio in groundwater. 2R model feasibility for chlorine predictions in bulk water have already been tested for surface waters (different types of water treatment) (Fisher et al., 2011). As per our knowledge none of the studies reported in literature has employed a 2R model for prediction of chlorine in groundwater. These results are important in the context of cities in developing countries where 50% of the water demand is met by groundwater pumping

(Grönwall et al., 2010). The groundwater is pumped to overhead tanks, chlorinated and is supplied through piped water connection. Employing models that accurately predict chlorine decay in surface water may not always be suitable for groundwater. Therefore water managers should be careful while employing similar models for predicting chlorine decay in surface water and groundwater.

4 Discussion

- ²⁰ Various studies have previously demonstrated applicability of 2R model for chlorine prediction in bulk water (surface waters), commencing from ICC 1 to 4 mg L⁻¹. The model was calibrated and validated for surface water that has undergone different types of water treatment/no treatment (Fisher et al., 2011; Rossman, 2006). In this study, an attempt was made to estimate optimal parameters for surface water and groundwater
- ²⁵ by calibrating 2R model to the experimental dataset. First, we calibrated 2R model against the entire range of chlorine decay dataset and estimated optimal parameters for surface water and groundwater. The 2R model estimated chlorine decay over time





and over the usual operating ranges of initial chlorine levels $(1-5 \text{ mg L}^{-1})$. Second, we validated the 2R model using two datasets; one from the complete chlorine decay dataset and the second from the dataset specifically reserved for validation.

For surface water, we obtained good model prediction, explaining 98% of the variance in data for all cases. 2R model provided an accurate representation of the effect of initial chlorine levels on chlorine decay in bulk water using a single set of invariant parameters. These results in conjugation with a sufficiently accurate prediction of wall decay will enable water management agencies to determine the ICC that allows residual targets at system extremities to be met.

- ¹⁰ For groundwater, 2R model slightly over (0 < t < 20 h) and under (40 < t < 60 h) estimated chlorine decay for ICC – 2 mgL^{-1} , raising questions about its suitability for chlorine prediction in groundwater. We found that the chlorine reaction in groundwater is different from that of surface water as slow reacting species dominate over fast reacting species in groundwater. This could be attributed to the presence of high levels of
- ¹⁵ manganese (inorganic component) and aromatic hydrocarbons in groundwater. Such groundwater when pumped out would have high slow to fast reacting species ratio. This characteristic of groundwater reduces chlorine consumption and thus will help in maintaining high levels of residual chlorine over long distances within the water distribution network.

20 5 Conclusions

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Through this study, first we estimated the fast and slow reacting components in test water and second we tested the feasibility of 2R model in predicting residual chlorine in bulk water from different sources i.e. surface water (river) and groundwater (deep aquifer). The 2R model was calibrated and validated using AQUASIM software. The experiments were performed with four chlorine decay datasets. The study came up with three important findings (a) we found that the ratio of slow to fast reacting components in groundwater was thirty times greater than that of the surface water. This



observations supports existing literature which indicates presence of high levels of slow reacting fractions (manganese and aromatic hydrocarbons) in groundwater, (b) 2R model accurately predicted chlorine decay in surface water as 98 % of the variance in the chlorine decay test was explained by the model and (c) in case groundwater, 2R model prediction accuracy declined at low ICC levels, only 87 % variance in data was explained by the model, which could be attributed to high slow to fast reactant ratio in groundwater.

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Table 1. Fast and slow reacting components present in different types of water.

Compounds	Organic	Inorganic
0	Aliphatic hydrocarbons Aromatic hydrocarbons	Nitrates, Sulphates, Ammonia Nitrites Manganese Mn (II)





Table 2.	Characteristics	of	data	set.
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Source	Treatment	ICC range (mg L^{-1})	Number of experimental runs	Temperature range (°C)
	Conventional treatment	1–5	4	25–30
Ground water	No treatment	1–5	4	25–30



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Table 3. Water quality parameters of test wate	er.
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Parameter	Surface water $(n = 3)$	Standard deviation	Groundwater $(n = 3)$	Standard deviation
рН	7.19	0.47	6.75	0.01
Conductivity (μ S cm ⁻¹)	434	30	1150	17
Nitrates (mgL ⁻¹)	3	1	177	20
Hardness(mgL ⁻¹)	180	56	352	8
Alkalinity (mg L^{-1})	196	4	165	3



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Table 4. Optimal parameter values and associated chi-squared values for the calibration dataset.

Source	$ICC \ (mg \ L^{-1})$	$C_{\rm S0}~({\rm mgL^{-1}})$	$C_{\rm f0}~({\rm mgL^{-1}})$	$K_{\rm f}$ (Lmg ⁻¹ h ⁻¹)	$K_{\rm s}~({\rm Lmg^{-1}h^{-1}})$	χ²	R^2
Surface water	1–5	2.88	0.51	2.55	0.01	0.18	0.978
Ground water	1–5	0.67	0.003	8.07	0.0069	0.48	0.998

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Table 5. Comparison of optimal parameters values obtained for different test waters.

Source	Treatment type	ICC range (mgL ⁻¹)	$C_{\rm f0}~({\rm mgL^{-1}})$	$C_{S0} (mgL^{-1})$	$K_{\rm f} ({\rm Lmg^{-1} h^{-1}})$	$K_{\rm s}~({\rm Lmg^{-1}h^{-1}})$	$C_{\rm S0}/C_{\rm f0}$	χ²	R^2
Surface water ^a	Conventional treatment	1–5	0.51	2.88	2.55	0.0069	6	0.18	0.978
Groundwater ^a	No treatment	1–5	0.003	0.67	8.07	0.013	223	0.48	0.998
Surface water ^b	Conventional treatment	1–4	0.808	3.88	0.261	0.0102	5	0.18	0.992
Surface water ^b	No treatment	1–4	0.761	2.69	0.199	0.0066	4	0.97	0.987
Surface water ^b	Not known	1–4	0.917	1.98	6.18	0.085	2	0.84	0.987

^a Bench scale test (our study). ^b Bench scale test (Fisher et al., 2011).











Figure 2. Chlorine decay in surface water and groundwater: markers - measured chlorine values; curves-values simulated by 2R models.

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Figure 3. Validation of 2R model for surface water and groundwater at different ICC.



