MODELLING CHLORINE DECAY IN DRINKING WATER SUPPLY SYSTEMS

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Abstract: The current work focuses on the analysis of flow hydraulics conditions effect on chlorine residual decay and on the development of a chlorine residual model in a water supply system from Águas do Algarve utility using EPANET 2. The work involves a literature review about the factors that influence chlorine decay and decay modelling, experimental studies on a helical pipe rig (friction factor analysis and the study of flow velocity on chlorine decay) and chlorine residual modelling of Águas do Algarve subsystem. This thesis contributes to a better understanding of the effect of flow hydraulics conditions on chlorine bulk decay rates and shows that these rates significantly increase with the flow velocity in turbulent flow for treated waters. An empirical formulation of decay rate that includes the flow velocity has been developed and can be incorporated in water quality simulators. The simulation of Águas do Algarve water supply system with first and nth order bulk decay kinetics had similar level of accuracy. However, results have shown that calibration and validation carried out only based on free chlorine analyzer measurements is not sufficient. Field measurements are required to calibrate and validate the models.

Keywords: chlorine residual modelling, chlorine decay, drinking water quality, water distribution systems.

1 INTRODUCTION

Chlorine residual is used worldwide as a hygienic barrier in drinking water systems. Chlorine concentration decreases as the water travels throughout the systems, where its levels must be enough to assure the disinfectant effectiveness. As the formation of toxic disinfection by-products increases with chlorine concentration, this must be maintained in drinking water within a quite narrow range, usually between 0.2 and 1.0 mg/L (WHO, 2011) and 0.2 and 0.6 mg/L according to Portuguese law (Decreto-Lei n.º 306/2007 de 27 de Agosto). However, the Annual Report of the Portuguese Water and Waste Regulator reveals that 45% of analysed samples have a chlorine residual insufficient to ensure hygienic barrier.

Chlorine decay models are important tools for the management of disinfectant concentration in drinking water systems, particularly for dosage optimization and chlorination facilities location. Despite the considerable amount of research developed in the last twenty years (Clark, 2011; Powell et al., 2000a; Rossman et al., 1994), chlorine decay phenomena and the factors that influence them are still being studied. Most water quality simulators describe chlorine decay by the sum of two groups of chemical reactions in which chlorine is consumed: reactions in the water (bulk decay) and at the pipes' internal surface (wall decay). Bulk decay is due to reactions of chlorine with compounds dissolved in the water, mainly those composing the Natural Organic Matter (NOM) and, to a minor extent, with inorganic compounds, like reduced iron and manganese forms (Powell et al., 2000a). The most widely used kinetic models in water supply systems simulation is the first order model (Clark & Sivaganesan, 2002):

$$\frac{dC_{Cl}}{dt} = -K_b C_{Cl} \tag{1}$$

where C_{Cl} is chlorine concentration, t is time, and K_b is the bulk decay coefficient (or bulk reaction rate constant). The nth order bulk decay kinetic model with

respect to chlorine is described by the following equation (Powell *et al.*, 2000b):

$$\frac{dC_{Cl}}{dt} = -K_b C_{Cl}^{\ n} \tag{2}$$

where the n is the order of the reaction with respect to chlorine. The decay coefficient is commonly determined through the laboratory bottle test (Powell *et al.*, 2000a).

Wall decay is particularly important in systems with metal pipes (Hallam *et al.*, 2002) owing to chlorine consumption in corrosion processes. Biofilm and sediments may also play a significant role in chlorine decay at the pipes' wall. Except in corroding iron pipes, bulk decay is responsible for most of chlorine consumption, having the wall demand a much smaller contribution to the overall chlorine decay (Kiene *et al.*, 1998).

Wall decay may also be described by a first order kinetic model (Rossman *et al.*, 1994):

$$\frac{dC_{Cl}}{dt} = -\frac{4k_f k_w C_{Cl}}{D(k_f + k_w)}$$
(3)

where k_w is the wall decay coefficient (or wall reaction rate constant), k_f is a mass transfer coefficient (velocity dependent) and D is the pipe diameter. The wall kinetic coefficient cannot be experimentally determined and is usually calibrated in order to fit calculated to measured chlorine concentrations.

Chlorine decay rates depend on the system operational conditions, such as water temperature, initial chlorine concentration, on the type and concentration of NOM (Brown *et al.*, 2011; Hallam *et al.*, 2003; Powell *et al.*, 2000a) and flow velocity (Hallam *et al.*, 2002; Menaia *et al.*, 2003).

An accurate chlorine decay model requires a suitable kinetic model (Fisher *et al.*, 2011; Powell *et al.*, 2000b) that is able to describe chlorine decay as a function of the influencing factors.

The temperature effect on chemical reaction rates coefficients is generally described by the empirical Arrhenius equation:

$$k = A e^{-E_a/(RT)} \tag{4}$$

where k is the reaction rate coefficient, A is the frequency factor, E_a is the activation energy, R is the ideal gas constant (8.31 J mol⁻¹ K⁻¹) and T is the temperature (Monteiro *et al.*, 2014). The Arrhenius parameters (A and E_a) are specific of each chemical reaction.

An increase in bulk decay coefficient with Reynolds number was reported by Menaia *et al.* (2003) and Ramos *et al.* (2010).

According to Menaia *et al.* (2003), water flow velocity effect on first order chlorine bulk decay coefficient can be described by:

$$K_b^d = K_b \left(1 + bUD \right) \tag{5}$$

Where K_b^d is the bulk decay coefficient at velocity U , K_{b} is the bulk decay coefficient at stagnant conditions and b is an adjustable parameter (103.20 m⁻² s for the tested conditions). Ramos et al. (2010) also observed a significant increase of parallel first order kinetic constants with increasing Reynolds number and described it with a linear function. The parameters in Equation 5 are related to tested waters temperature and NOM reactivity towards chlorine, thus hindering a straightforward implementation of these equations in chlorine residual modelling. Additionally, the testing conditions used by Menaia et al. (2003) and Ramos et al. (2010) do not reflect real conditions in drinking water networks. Those authors used commercial humic acids as surrogate for NOM at relatively high concentrations (5 mg C/L as Total Organic Carbon).

The aim of this work is the study of the effect of hydraulic conditions on chlorine bulk decay coefficient in treated waters and to develop an empirical formulation for K_b as a function of hydraulic parameters. Additionally, this work intended to contribute to the implementation of residual chlorine modeling in a Portuguese water transmission system

and to identify the major difficulties and uncertainties in water quality modelling.

2 EXPERIMENTAL SETUP AND PROCEDURES

Two sets of experiments were performed in a laboratory pipe rig. The first tests aimed at determining the flow velocity in the pipe rig for the different operation conditions for pipe friction analysis. The second set of tests was performed in order to analyse the effect of hydraulic conditions on chlorine decay bulk coefficient.

2.1 Pipe rig description

The pipe rig (Figure 1), assembled in the Laboratory of Hydraulics and Environment of Instituto Superior Técnico, is a helical pipe closed loop of high-density polyethylene (HDPE) with about 100 m long and 32 mm diameter. The system was about 2 m high and included a recirculation pump (Filtra N 24D, KSB) connected to a variable-frequency drive, a 4 L capacity standpipe on top, a sampling port, a 15 cm long transparent polyvinyl chloride (PVC) pipe branch and a valve on the bottom to fill and to drain the system. All fittings were made of PVC and the pump internal materials were plastic.



Figure 1 – Pipe rig.

The system operates in a closed loop and reproduces a hydraulic pressure system consisting of a long pipe without branching.

2.2 Flow velocity estimation

Tracer tests were performed at 17 flows with demineralized water. For each test, sodium chloride was injected with a syringe at the beginning of the HDPE pipe. The conductivity was continuously measured with a conductivity probe placed at the end of the HDPE pipe. The tracer time between the injection section and the conductivity probe was measured, as well as the water temperature. The piezometric head variation between two sections in the pipe was measured using two piezometers installed in the circuit.

2.3 Chlorine decay tests

For each experiment, 80 L of Tavira Water Treatment Plant final water were chlorinated with sodium hypochlorite (ca. 1.0 mg/L) in a plastic tank and immediately pumped into the pipe rig until the system was completely filled with water. Trapped air was thoroughly eliminated. When no air bubbles were observed, water samples were collected for initial chlorine concentration measurement and to completely fill twelve amber glass 100 mL bottles. Then, loop water flow velocity was set (Table 1) and kept steady for several days. Chlorine concentration in the pipe rig water was monitored by collecting and analysing samples with the N,N-diethyl-p-(DPD) phenyldiamine method, using а spectrophotometer (Dr. Lange, Cadas 50). The water monitored temperature was with а digital thermometer inserted in the pipe. The bottles were placed in a cardboard box, to protect from light, next to the pipe rig at the same ambient temperature. Chlorine concentration and temperature in the bottles water were measured at the same time intervals as for the pipe rig samples. Tests lasted until chlorine concentration in rig pipe water decreased to bellow the method measurement limit (0.1 mg/L) or for 7 days. Experiments comprised five different flow rates for tested water (Table 1). One additional experiment with chlorinated demineralized water was performed at 1.07 m/s to evaluate pipe's wall contribution to overall chlorine decay. Decay tests were done at Reynolds numbers within the range from 4089 to 16160, hence included both laminar and turbulent flow regimes.

3 FRICTION RESISTANCE IN HELICAL PIPES

3.1 Friction factors

In an incompressible steady flow, piezometric head variation between two sections in the pipe it is the head loss between those sections. The unit headloss (J) for a pipe with constant cross section can be determined by Darcy-Weisbach equation:

$$J = f \frac{U^2}{2gD} \tag{6}$$

where f is the Darcy friction factor, g is the gravity acceleration and D is the pipe inner diameter. The friction factor formulation varies with flow regimes and pipe configuration. In laminar flows in straight pipes, Hagen-Poiseuille law can be used:

$$f = \frac{64}{\text{Re}} \tag{7}$$

where Re is the Reynolds number. In turbulent flows in smooth straight pipes, the Karman-Prandtl semiempirical formula is used:

$$\frac{1}{f^{0.5}} = -2\log\left(\frac{2.51}{\text{Re}\,f^{0.5}}\right) \tag{8}$$

Ito (1959) has study the friction factors formulas for flows in helical pipes. From his results, the friction factor for laminar flow is given by:

$$f = \frac{64}{\text{Re}} \frac{21.5De}{\left(1.56 + \log De\right)^{5.73}} \tag{9}$$

and for turbulent flow is given by:

$$f = 0.304 \,\mathrm{Re}^{-0.25} + 0.029 \left(\frac{D}{d_c}\right)^{0.5}$$
 (10)

where d_c is the curvature diameter and De is the Dean number ($De = \operatorname{Re}(D/d_c)^{0.5}$).

In helical pipes, the laminar to turbulent flow transition occurs at a higher Reynolds number. Srinivasan *et al.* (1970) indicate that the flow transition occurs around the critical Reynolds number, Re_{c} :

$$\operatorname{Re}_{c} = 2100 \left[1 + 12 \left(\frac{D}{d_{c}} \right)^{0.28} \right]$$
 (11)

3.2 Experimental friction factor formulations

The specific Re_c of this pipe rig is 6140 (for D = 0.0264 m and $d_c = 1.027$ m).

The experimental friction factor for 17 tests of the first set were calculated and were compared with existing formulations for laminar (Figure 2) and for turbulent flows (Figure 3).







Figure 3 – Comparison of existing formulations and experimental results for laminar flow.

Results have shown that, in helical pipes, the friction is higher than in straight pipes for both laminar and turbulent flows. However, the experimental results of the friction factor for Re < 11 000 (in turbulent flow) were lower than the ones for straight pipes.

Empirical formulations for friction factor of the pipe rig flow were developed for laminar flow:

$$f = 22.246 \,\mathrm{Re}^{-0.78} \tag{12}$$

and for turbulent flow:

$$f = 0.1115 \,\mathrm{Re}^{-0.14} \tag{13}$$

3.3 Flow velocity estimation

An empiric formulation were determined to estimate the flow velocity in the pipe rig with unit headloss (J) through the Equations (12) and (13).

The flow velocity for laminar flow can be estimated by:

$$U = \left(\frac{2gJD^{1.78}}{22.246\nu^{0.78}}\right)^{0.82}$$
(14)

and for turbulent flow by:

$$U = \left(\frac{2gJD^{1.14}}{0.1115v^{0.14}}\right)^{0.54}$$
(15)

The unit headloss (J) is determined by the ratio of the measured piezometric head variation in two piezometers installed in the pipe rig and the distance (L) between them.

4 HYDRAULIC CONDITIONS EFFECT ON CHLORINE DECAY

Decay test with demineralized water (blank tests) showed only minor decay of chlorine concentration through time, both in the pipe rig and in the bottles. A simple first order kinetic model was used to describe chlorine decay in demineralized water bottle tests and estimate bulk decay coefficient. Chlorine decay in the pipe rig in the blank test was modelled using first order K_b (0.0015 h⁻¹) and calibrating the wall decay coefficient by minimizing the sum of the squared residuals between measured and modelled values. A

first order kinetic model was assumed for wall decay. A rather low value of 2.6x10⁻⁰⁶ m/h was found for k_w , which is in accordance with Clark (2011), who states the low contribution of plastic pipes for chlorine decay. A n^{th} order kinetic model (n = 2) was used and accurately described chlorine decay in the bottles with the tested water (Table 1). Coefficient of determination (R2) and Root Mean Squared Error (RMSE) assessed goodness of fit of the model to experimental data. Determined bulk decay coefficients varied between 0.020 and 0.067 L/mg/h due to differences in water temperature and possibly due to variations in water quality. At the smallest water flow velocity tested (0.15 m/s), chlorine decayed in the pipe at the same rate as in the bottles. For all the other tested velocities, chlorine decayed faster in the pipe rig, which is in accordance with Menaia et al. (2003) and Ramos et al. (2010) findings.

Chlorine decay in the pipe rig in each experiment was modelled by the usual "bulk + wall" approach and using K_b from bottle tests and previously determined k_w from blank test:

$$\frac{dC_{Cl}}{dt} = -K_b C_{Cl}^2 - \frac{4}{D} \frac{k_f k_w}{(k_f + k_w)} C_{Cl}$$
(16)

The obtained RMSE (Table 1) is relatively high (above 0.06 mg / L) and shows a tendency to increase with water flow velocity, as higher values were obtained with higher tested velocities (0.52 and 0.61 m/s). Apparently, there is an increasing inability of the model to describe chlorine decay in the pipe as the water flow velocity increases. Hence, a new modelling approach was developed in order to incorporate flow velocity effect on K_b . Bulk decay coefficient in Equation (16) was replaced by an analogous coefficient, determined in dynamic flow conditions, K_b^d :

$$\frac{dC_{Cl}}{dt} = -K_b^d C_{Cl}^2 - \frac{4}{D} \frac{k_f k_w}{(k_f + k_w)} C_{Cl}$$
(17)

The new coefficient was determined by fitting Equation (17) to experimental results of each test. In order to

compare the obtained values with K_b from bottle tests, the ratio of the two coefficients was computed (Table 2). Results have shown that the ratio increased with flow velocity (Figure 4) and that bulk decay coefficient in turbulent flow conditions may double its value. These results are in agreement with Menaia *et al.* (2003) and Ramos *et al.* (2010) who observed an increase in bulk decay coefficient with flow velocity and Reynolds number, respectively.

A linear relationship between the ratio K_b^d/K_b and Reynolds number (Figure 4) was also developed:

$$K_b^d = K_b \left(1 \times 10^{-4} \,\mathrm{Re} + 0.62 \right) \tag{18}$$

A similar equation was derived between the ratio K_b^d/K_b and flow velocity:



Figure 4 – Ratio K_b^d/K_b variation with Reynolds number. Equations (18) and (19) describe hydraulic conditions effect on bulk decay coefficient in the test rig. The equations' parameters are probably a characteristic of the tested water and of the pipe system configuration.

	Bottle test			Pipe rig test				
Test	K _b	RMSE	R ²	U	k_{f}	RMSE	R ²	Т
	(L /mg /h)	(mg/L)		(m/s)	(m/h)	(mg/L)		(°C)
1	0.057	0.09	0.95	0.15	0.042	0.06	0,98	18.0
2	0.067	0.05	0.99	0.34	0.082	0.10	0,95	25.2
3	0.061	0.06	0.97	0.43	0.101	0.11	0,93	21.4
4	0.042	0.05	0.98	0.52	0.120	0.17	0,80	23.7
5	0.020	0.05	0.98	0.61	0.139	0.14	0,83	20.1
6	0.0015*	0.035	0.83	1.07	0.227	0.036	0.88	20.0

Table 1 – Determined coefficients for each test and goodness of fit of the bulk + wall modelling approach.

Note: first order decay coefficient in 1/h

Table 2 – Estimated dynamic bulk decay coefficient, goodness of fit of the proposed model and comparison with bulk decay coefficient under static conditions.

Test	U_{\parallel}	Re	K_{b}^{d}	RMSE	R ²	K_b^d / K_b
	(m/s)		(L /mg /h)	(mg/L)		
1	0.15	4089	0.055	0.06	0.98	0.96
2	0.34	8853	0.098	0.07	0.97	1.46
3	0.43	11224	0.105	0.05	0.99	1.72
4	0.52	13686	0.095	0.04	0.99	2.26
5	0.61	16160	0.039	0.04	0.99	1.95

5 CASE STUDY

5.1 Case-study description

The case study was carried out in a sector of the drinking water transmission system that supplies eastern Algarve, Portugal. It comprises a 23 km long,

large-diameter trunk main with 7 delivery points. At each point, water is delivered to service storage tanks that are managed by municipal water utilities. The system is supplied by the Tavira Water Treatment Plant (WTP) and carries water to Cabeço service tank at the downstream end (Figure 5). Pipe diameters range from 450 and 1500 mm in the main line, with delivery branches ranging from 100 to 400 mm. Water flows by gravity and unidirectional. Flow is controlled by water levels in the tanks and, therefore, depends on the demand patterns at the delivery points. Water is treated by a conventional process for superficial waters consisting of pre-oxidation with ozone, followed by coagulation/flocculation/sedimentation, sand filtration and final disinfection with gaseous chlorine. Average chlorine content was 0.98 mg/L at the WTP outlet and average water temperature was 13°C during the study period. Water had relatively low organic (1.3 mg C/L as total organic carbon) and inorganic contents (iron, ammonia and manganese concentrations below detection limits) and therefore, low chlorine demand is expected. Predominant pipe materials are ductile iron with aluminous cement lining and steel. Average infrastructure service age is about 15 years.



Figure 5 – Case study's EPANET model.

5.2 Methodology

The hydraulic model was implemented in EPANET 2.0. Water consumption patterns at the nodes were developed based on water flow measurements at each of the seven delivery points, which were obtained from the telemetry system, for a 10 days period (18th to 27th January 2012). Time step of 1 minute was used for hydraulic simulation. For water quality modelling, three patterns of chlorine concentration were developed based on measured concentrations by online analyzers located at the WTP outlet and at two delivery points (Perogil and Santa Rita). All data wer registered at 1 min time interval and checked for outliers and consistency. Chlorine concentration measured at the WTP outlet was set on EPANET as the only source of chlorine in the system. The other two series of chlorine concentration were used for the model's calibration (data from 21st to 24th January 2012) and validation (data from 25th to 27th January 2012).

A sensitivity analysis of water quality simulation time step was performed in order to evaluate how much this parameter may affect the accuracy of the modelling and to estimate the appropriate value to be used in chlorine decay simulations. For such purpose, water age at Perogil, Santa Rita and Cabeço were computed using quality time steps between 0.25 and 60 min. Assuming that the most accurate water age is the one computed using the smallest quality time step (Georgescu & Georgescu, 2012), mean relative errors were computed and compared.

A previous study on chlorine bulk decay kinetics was performed (Monteiro *et al.*, 2014) for Tavira treated water in winter season and at several temperatures. Two decay models were selected (simple first order and nth order with *n* of 1.2) and tested in chlorine decay modelling in the transmission system. The bulk decay coefficients at the average water temperature in the system were 0.27 day⁻¹ for first order model and 0.35 $L^{0.2}/mg^{0.2}/day$ for nth order model.

Chlorine residual in the transmission system was firstly simulated assuming that only bulk decay was occurring. Then, a wall decay coefficient was iteratively calibrated by minimizing the sum of the squared differences simulated between and measured chlorine concentrations at the two nodes where online analyzers were located. Wall decay was modelled assuming a first order model, as the pipe materials are predominantly of low reactivity (lined ductile iron). A single wall coefficient was calibrated for the whole system since over 85% of the pipes are of the same material and of identical service age.

5.3 Water age sensitivity analysis

Results have shown that water age relative errors significantly increase with quality time step (Figure 6) for the three locations. Relative error was about 6 to 10% when quality time step was set to 5 min, which is the recommended value on EPANET manual (Rossman, 2000). When the quality time step is set to 1 min or less, low mean errors of approximately 1% (Figure 6) were obtained. Therefore, considering the best compromise between simulation time and accuracy of the model, all henceforward chlorine decay simulations were performed using 1 min as quality time step. These results have shown that the Lagrangian time-driven simulation method used by EPANET is sensitive to the calculation step and that the choice of quality time step is extremely important when implementing a water quality model.



Figure 6 –Water age relative error for each tested quality time step at Perogil, Santa Rita and Cabeço delivery points.

5.4 Chlorine decay modelling

Modelling chlorine residuals assuming that the pipes' walls would not have a significant demand has led to poor correlations (R² less than 0.85) between computed and measured chlorine concentrations at Perogil and Santa Rita delivery points, whichever bulk decay kinetics were used (Figure 7a). In these simulations, both models overestimated chlorine concentrations and only a minor difference is noticed between the two tested models. These results suggest that, on one hand, bulk decay is probably not being accurately described, since flow velocity effect was not taken into account in the modelling, and, on the other hand, that wall demand is an important part of chlorine decay in this system and must be incorporated in the modelling as well.

By calibrating the wall decay coefficient, better correlations were obtained (R² of about 0.93) between measured and computed values, as observed by Vasconcelos et al (1997) (Figure 7b). Calibrated k_w were 0.035 and 0.022 m/day when using bulk decay kinetics of first and nth order, respectively. Estimated wall decay coefficient was higher when the first order kinetic model was used for bulk decay description, which is due to the higher discrepancies between this model's computed chlorine concentrations and measured ones. This suggests that uncertainties in bulk decay simulations are being partially incorporated in the calibrated wall decay coefficient, thus in accordance with Fisher et al. (2011) findings. RMSE of the 0.03 mg/L whichever bulk decay kinetics are used, which is lower than the precision of the most widely used chlorine concentration measurement method (0.05 mg/L for HACH colorimeters). Therefore, the models were considered sufficiently accurate.





When comparing overall chlorine predicted concentrations with the measured ones at Perogil by calibrated models were about the online chlorine analyzer over time (Figure 8), it is noticed that the models accurately described chlorine concentration during the calibration period but not as well in the validation one. The models were able to simulate chlorine peaks at the exact times they were detected by the online analyzer, thus denoting that the hydraulic model was well calibrated.

However, it is observed that the models predicted much lower chlorine concentrations between 140 and 150 h of simulation time than the measured concentrations. This is probably because the models were based on a chlorine concentration time pattern that was built with WTP outlet analyzer measurements. The models were, therefore, vulnerable to possible incorrect measurements of this analyzer, although all frequently calibrated. These analyzers were uncertainties make it difficult to understand whether the discrepancies were due to the models inability to describe chlorine decay in the system or due to incorrect chlorine measurements, at Perogil or at the WTP. Hence, model validation, as well as chlorine concentration time pattern set up, should not rely only on online analyzers but also on field sample measurements, carried out at several locations in the system and over the study period.



Figure 8 – Comparison of computed and measured chlorine concentration over the simulation period at Perogil.

6 CONCLUSIONS

Chlorine bulk decay coefficient of treated water significantly increases with water flow velocity for turbulent conditions. A linear relationship between bulk decay coefficient at turbulent conditions and Reynolds number was developed. Such expression might be incorporated in chlorine decay models, making use of EPANET-MSX potential. Using k_b from bottle tests for modelling chlorine in water supply systems is likely to result in models of low accuracy. Including the effect of water flow velocity will reduce the importance of wall demand on overall chlorine decay.

When modelling chlorine residual in water supply systems, relying on online analyzer's measurements can be of great advantage, although these data must be validated and complemented with field sample measurements, particularly at the source point of chlorinated water in the system. Additionally, for greater accuracy, chlorine residual simulations on EPANET must be performed using small quality time steps.

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