RESEARCH ARTICLE

Chlorine fate and transport in drinking water distribution systems: Results from experimental and modeling studies

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Abstract It has become generally accepted that water quality can deteriorate in a distribution system through microbiological and chemical reactions in the bulk phase and/or at the pipe wall. The most serious aspect of water quality deterioration in a network is the loss of the disinfectant residual that can weaken the barrier against microbial contamination. Studies have suggested that one factor contributing to the loss of disinfectant residuals is the reaction between bulk phase disinfectants and pipe wall material. Free chlorine loss in corroded metal and PVC pipes, subject to changes in velocity, was assessed during an experiment conducted under controlled conditions in a specially constructed pipe loop located at the US Environmental Protection Agency's (EPA's) Test and Evaluation (T&E) Facility in Cincinnati, Ohio (USA). These studies demonstrated that in older unlined metal pipes, the loss of chlorine residual increases with velocity but that wall demand in PVC was negligible.

Keywords chlorine fate, transport, drinking water

1 Introduction

A number of factors can influence water quality in distribution systems including: chemical and biological quality of source water (Clark and Coyle, 1990). A serious public health impact of water quality deterioration is the loss of disinfectant residual, which can weaken the barrier against microbial contamination. Internal corrosion and deterioration of the pipe wall material can be the result of physical action that erodes the lining or surface coating of a pipe, chemical dissolution that leaches a pipe lining or wall material, or electrochemical reactions that remove metal from the wall of the pipe.

There have been some reports that the loss of chlorine residuals in corroded unlied metallic pipes increases with increasing velocity (Powell, 1998; Grayman et al., 2002; Doshi et al., 2003). Powell (1998) observed significant differences in wall demand between unlined metallic and PVC pipe.

To assess the wall demand for free chlorine in various pipe materials (PVC and unlined ductile iron) subject to changes in velocity, a study was conducted under controlled conditions in specially constructed pipe loops (distribution system simulator) located at the US Environmental Protection Agency's (EPA's) Testing and Evaluation (T&E) Facility in Cincinnati, OH (USA) (Clark and Haught, 2005).

The PVC pipe exhibited a much lower wall demand than unlined ductile iron pipe and it was found that there are some weaknesses in the various modeling approaches commonly proposed. It was concluded that these transport models should be applied with some degree of caution.

2 Previous studies

Many studies have attempted to determine the factors that cause the deterioration of water quality in networks (Williams, 1958; Clark, et al., 1991; Clark et al., 1994; Benjamin et al. 1996; Vasconcelos et al., 1997). In a comprehensive study of chlorine residual losses in drinking water distribution systems, Powell et al. (1998) found that cast-iron pipe typically has 10 to 100 times higher wall demand than PVC pipe. He also found an increase in wall demand with velocity which was most prominent in cast-iron pipe. A comprehensive field study by the Detroit Water and Sewerage Department, showed that higher flow rates resulted in greater residual chlorine loss in the pipe (Grayman et al., 2002; Doshi et al., 2003).

Digiano and Zhang (2005) used a bench-scale reactor to show that chlorine decay rate was described by zero-order reaction kinetics for cast-iron pipe. The zero-order rate was

larger at higher velocities because of higher mass transfer to the pipe surface, although, a limit was reached. For the ductile-iron pipe, the chlorine decay kinetics were found to be first-order with respect to chlorine concentrations. Al-Jasser (2007) found that the effect of pipe age was most evident in cast-iron pipes whereas steel pipes were less affected.

A recent study conducted by Clark et al. (2010) has confirmed that increased velocities in unlined pipes can result in increased losses in chlorine residual. However, increased velocities have no effect on chlorine residual losses in PVC pipes.

3　Data collection procedures

All experimental data were collected according to EPA Standard Methods in two recirculating pipe loops located at EPA's T&E Facility in Cincinnati, Ohio (Rossman et al., 2001). These pipe loops are described in detail elsewhere, and the flow characteristics were evaluated using tracer testing (Panguluri et al. 2007) and are part of a Distribution System Simulator (DSS). The DSS consisted of two sets of three individual 15.24 cm (6 inches) diameter pipe loops (a total of six loops). The three loops in each set are stacked above each other on a project pad. Each loop is 27 m (88 feet) in length and consists of 15.24 cm (6 inches) pipe of various materials.

4　Experimental protocol

Two of the six DSS loops were used in this study (Loops 4 and 5) in a continuously circulating "batch" mode (no outflow and no inflow). Loop 4 is unlined ductile-iron pipe, and Loop 5 is made of PVC. Five experimental runs were conducted in each loop in an attempt to characterize the loss of chlorine in each loop as a function of velocity. The tests were performed at the following constant recirculation flow rates: 0.011 m^3/min (3 gpm); 0.023 m^3/min (6 gpm); 0.038 m^3/min (10 gpm); 0.379 m^3/min (100 gpm); and zero flow or stagnant conditions. Water temperature ranged between 13.09°C (55.6°F) and 23.72°C (74.7°F), and the pH of the water ranged between 8.19 and 9.06. The initial free chlorine concentrations were spiked at approximately 2.5 to 2.6 mg/L and allowed to dissipate during the experiment.

The following parameters were collected as grab samples in the test loops and the reservoir (9.46 m^3 (2500 gal) stainless steel mixing tank) simultaneously: free chlorine, turbidity, temperature (in-line), pH (in-line) in the loop, total organic carbon (TOC), THMS, and HAAs. No data were utilized from samples when the free chlorine concentration decreased below 0.05 mg/L. After several experimental runs, it was found that the reservoir parameters changed only slightly during an experimental run, therefore reservoir sampling was conducted on a reduced schedule. Tables 1 and 2 summarize the average temperature, pH, turbidity, TOC, HAA9s, and THMs for the experiments, although these values varied over time.

5　Experimental results

The source water for this set of experiments was GAC treated distribution system water. For purposes of this analysis, the following is assumed:

Table 1 Summary of average values for Pipe Loop 4 (unlined ductile-iron) physical and chemical parameters (chlorine concentration of 2.5 to 2.6 mg/L)

Flow rate[a]/(m$^3 \cdot$ min^{-1}) (gpm)	Facility	Turb[b]/NTU	Temp[b]/°C(°F)	pH[b]	TOC[def]/(mg$\cdot$ L^{-1})	THMs[cdef]/(μg$\cdot$ L^{-1})	HAA9s[def]/(μg$\cdot$ L^{-1})
(1)	(2)	(3)	(4)	(5)	(6)	(7)	(8)
0.000 (0)	Reservoir	0.43	23.52 (74.3)	8.55	0.86	33.21	46.4
	Pipe Loop	0.72	23.72 (74.7)	8.75	1.00	64.7	58.3
0.011 (3)	Reservoir	0.63	22.31 (72.5)	8.53	1.13	33.12	131.7
	Pipe Loop	1.21	20.09 (68.2)	8.84	1.13	57.50	98.9
0.023 (6)	Reservoir	0.61	20.07 (68.1)	8.52	1.09	27.80	13.9
	Pipe Loop	0.67	19.46 (67.0)	8.85	1.15	48.69	26.4
0.038 (10)	Reservoir	0.52	18.67 (65.6)	8.56	0.66	32.63	44.0
	Pipe Loop	0.56	19.28 (66.7)	8.92	0.60	49.41	51.8
0.379 (100)	Reservoir	0.49	13.09 (55.6)	8.19	1.15	24.79	43.3
	Pipe Loop	0.54	17.94 (64.3)	8.81	1.25	46.40	55.6

Notes: a) The flow rate was recorded at the start of each test and monitored (but not recorded) at each sampling event. Flow rate is monitored by reading an in-line flowmeter. b) pH, temperature and turbidity were collected via grab sample in Loops 4 and 5 and from the reservoir. c) Critical THM components include chloroform, dichlorobromomethane, chlorodibromomethane, and bromoform. d) EPA. 1983. Methods for Chemical Analysis of Water and Wastes. Office of Research and Development, Washington, DC. EPA 600/4-79-020. e) EPA. 1990. Methods for the Determination of Organic Compounds in Drinking Water, Supplement I. Office of Research and Development. EPA 600/4-90-020. f) Standard Methods for the Examination of Water and Wastewater, 20th Edition, Method 4500-H

Table 2 Summary of average values for Pipe Loop 5 (PVC) physical and chemical parameters (chlorine concentration of 2.5 to 2.6 mg/L)

Flow rate[a]/(m³·min⁻¹) (gpm)	Facility	Turb[b]/NTU	Temp[b]/°C (°F)	pH[b]	TOC[def]/(mg·L⁻¹)	THMs[cdef]/(μg·L⁻¹)	HAA9s[def]/(μg·L⁻¹)
(1)	(2)	(3)	(4)	(5)	(6)	(7)	(8)
0.0 (0.0)	Reservoir	0.93	23.55 (74.4)	8.54	1.32	34.13	55.0
	Pipe Loop	0.47	24.29 (75.7)	8.56	1.01	48.9	62.0
0.011 (3)	Reservoir	0.38	22.94 (73.3)	8.58	0.97	35.38	73.0
	Pipe Loop	0.47	20.37 (68.7)	8.58	1.05	42.02	65.3
0.023 (6)	Reservoir	0.39	20.29 (68.5)	8.53	1.12	30.26	21.3
	Pipe Loop	0.38	19.30 (66.7)	8.57	1.12	36.94	17.3
0.038 (10)	Reservoir	0.27	17.78 (64.0)	8.50	0.79	34.07	42.7
	PipeLoop	0.55	31.94 (89.5)	8.55	0.58	39.47	48.5
0.379 (100)	Reservoir	0.56	15.11 (59.2)	8.39	1.28	30.34	43.3
	Pipe Loop	2.05	18.21 (64.8)	8.45	1.23	40.78	55.6

Note: a,b,c,d,e,f) with the same meanings as in Table 1

Table 3 Calculation of chlorine wall demand for ductile iron and PVC pipe

Flow rate /(m³·min⁻¹) (gpm)	Bulk demand /h⁻¹	DIP[a] total chlorine demand/h⁻¹	DIP[a] wall demand/h⁻¹	DIP regression coefficients	PVC total chlorine demand/h⁻¹	PVC wall demand/h⁻¹	PVC regression coefficents
(1)	(2)	(3)	(4)	(5)	(6)	(7)	(8)
0.0 (0.0)	0.042	0.116	0.074	0.942	0.027	−0.015[b]	0.929
0.011 (3)	0.042	0.172	0.130	0.944	0.014	−0.028[b]	0.660
0.023 (6)	0.042	0.245	0.203	0.901	0.011	−0.030[b]	0.757
0.038 (10)	0.042	0.396	0.354	0.924	0.013	−0.029[b]	0.857
0.379 (100)	0.042	0.454	0.412	0.934	0.019	−0.023[b]	0.893

Notes: a) Ductile iron pipe; b) assumed as zero

$$C_t = C_0 e - (k_b + k_r)t, \qquad (1)$$

where C_t = the concentration of chlorine at any time t; C_0 = the initial concentration of chlorine; k_b = the bulk decay constant; k_r = the wall decay constant; and t = time in seconds.

Taking the natural log of Eq. (1) and rearranging yields:

$$\ln\left(\frac{C_t}{C_0}\right) = -(k_b + k_r)t. \qquad (2)$$

A regression analysis was conducted in which the bulk decay in the tanks and reservoirs was regressed against time of decay, resulting in k_b of 0.042 h⁻¹ (1.17×10^{-5} s⁻¹), as shown in Table 3. The calculated k_b was compared to the total chlorine demand ($k_b + k_r$) for the Loop 5 experiments. It was found that all of the demand data from Loop 5 fell within one standard deviation of the bulk demand at all velocities, and in some cases, was less than the calculated bulk demand. Therefore, it was concluded that the wall demand for PVC was negligible and was assumed to be zero in this study.

In Fig. 1, the solid lines represent the demand over time (uncorrected for bulk decay) for PVC pipe and the dotted lines represent the demand over time (uncorrected for bulk decay) for unlined ductile iron pipe. It is obvious that the total chlorine demand ($k_b + k_r$) in the unlined metallic pipe is much greater than the total demand for the PVC pipe. It is also very clear from Table 3 that in the unlined metallic pipe wall demand increases dramatically with velocity.
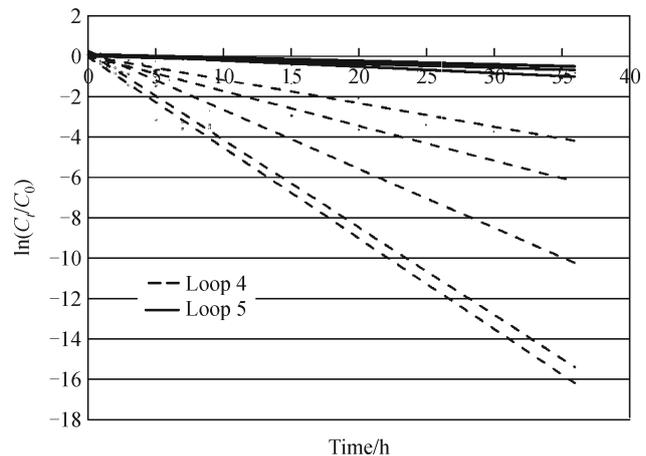

Fig. 1 $\ln(C_t/C_0)$ versus time in hours for Loop 4 (unlined metallic pipe) and Loop 5 (PVC) uncorrected for bulk demand

6 Model development

To examine the application of these data several models were explored based on work conducted by Biswas et al. (1993), and Rossman et al., (1994), and Clark and Haught (2005). For this analysis, steady-state conditions are assumed $\left(\dfrac{\partial c}{\partial t} = 0\right)$ which yields the equation for the steady-state transport of chlorine, shown below:

$$u\frac{dc}{dx} + k_b c = -\frac{2}{a} J_w, \qquad (3)$$

where c is the bulk chlorine concentration, u = the cross–sectional average flow velocity in cm/s; dc/dx = the total flux of chlorine in the axial direction, assumed to be a function of the axial position, x (cm) and time (s); k_b = the first–order bulk–decay rate for chlorine (c) in s^{-1}; α = the radius of cylinder in cm centered on the pipe axis; and J_w = the radial flux to the pipe wall in (mg·cm)/(L·s).

As mentioned, for purposes of this analysis, the following three modeling approaches were examined:
- Wall reaction limited model;
- Zero-order reaction limited;
- Mass transfer limited.

6.1 Wall reaction limited model

In the Wall reaction limited model, the equation assumed to describe the mass balance of chlorine that is transported to the wall, and the chlorine that is consumed at the pipe wall is:

$$J_w = k_{mt}(c - c_w), \qquad (4)$$

where k_{mt} is the mass transfer coefficient in cm/s; and c is the concentration in the bulk phase in mg/L and c_w is the wall concentration. This is the basic underlying assumption of the EPANET model.

Therefore, the expression used to describe the mass balance of chlorine that is transported to the wall, and the chlorine that is consumed at the pipe wall is

$$k_{mt}(c - c_w) = k_w c_w, \qquad (5)$$

where k_w is the wall demand coefficient in cm/s.

Under steady-state conditions and making the appropriate substitutions, Eq. (3) can be re-written as:

$$\frac{dc}{dt} = -k_b c - \left[\frac{2}{a}\right][k_{mt}k_w/(k_{mt} + k_w)]c. \qquad (6)$$

The total decay of chlorine can therefore be expressed in terms of a single first-order coefficient, K (s^{-1}) given by:

$$K = k_b + (2/a)\left(k_{mt}k_w/(k_{mt} + k_w)\right). \qquad (7)$$

In Eq. (7), the first term is the bulk decay rate, and the second term is the wall decay rate that consists of the wall demand coefficient (k_w); the mass transfer coefficient (k_{mt}); and the pipe radius (a). If we assume a constant k_w, the second term on the right hand side of Eq. (7) approaches zero as k_{mt} decreases and becomes asymptotic to $2k_w/a$ with increasing k_{mt}. The implications of this property were discussed in Clark and Haught (2005). Equation (7) can be rewritten as follows (Rossman et al., 1994):

$$k_w = \frac{k_{mt}\dfrac{a}{2}(K - k_b)}{k_{mt} - \dfrac{a}{2}(K - k_b)}. \qquad (8)$$

As can be seen under certain conditions, Eq. (8) can result in negative values for k_w.

Using standard calculations for mass transfer coefficient a value for wall demand (k_w) for the unlined metallic pipe (Loop 4) can be "back-calculated." These calculations are summarized in Table 4 and Fig. 2. As demonstrated by Eq. (8) for very low (laminar or transititional) flow rates, the wall demand parameter k_w can be negative.

As can be seen, the values for k_w vary with flow in contrast to the underlying assumption, embedded in EPANET, that the wall demand parameter is constant for a given length of pipe. This result is supported by previous studies (Clark and Haught, 2005; Clark et al., 2006) as well as by these data. It is apparent that the wall demand value in this model has no physical meaning (Clark et al. 1995, Clark et al. 2010). The implication from this analysis is that the reaction limited model should be used with caution.

Table 4 Summary of k_w (EPANET) and k_0 (zero–order reaction) wall demand calculations (Loop 4)

Flow rate		Velocity				K_{mt}	k_w	k_o
gal·min^{-1}	10^{-6} m^3·s^{-1}	/(cm·s^{-1})	Re	Sc	Sh	/(cm·s^{-1})	/(cm·s^{-1})	/(mg·cm·L^{-1}·h^{-1})
0.0	0.0	0.0	0.0	822.1	3.7	2.9219E−06	−2.9932E−06	0.146
3.0	189.3	1.0	1576.5	822.1	13.3	1.0618E−05	−1.1276E−05	0.274
6.0	378.5	2.1	3153.0	822.1	167.0	1.3369E−04	−2.7601E−04	0.604
10.0	630.9	3.5	5255.0	822.1	261.8	2.0958E−04	−4.1920E−04	0.975
100.0	6309.0	34.6	52549.9	822.1	1985.9	1.5898E−03	6.8860E−04	1.016

Fig. 2 EPANET wall demand constant (k_w) versus mass transfer coefficient

Fig. 3 Zero-order wall reaction coefficient versus mass transfer coefficient for unlined metallic pipe

6.2 Zero-order reaction

The Zero-order model is based on the assumption that the amount of chlorine (disinfectant) transported to the pipe wall is reaction limited, but that the radial flux of the disinfectant must equal the amount of chlorine that reacts at the pipe wall. Therefore, we would assume that the mass transfer equation is

$$k_{mt}(c - c_w) = k_0, \qquad (9)$$

where k_0 = the reaction constant; and $J_w = k_0$.
Substituting the relationship $J_w = k_0$ into Eq. (3) yields

$$u\frac{dc}{dx} + k_b c = -\frac{2}{a}k_0. \qquad (10)$$

The resulting equation is as follows:

$$c = c_0 e^{-k_b t} - \frac{2k_0}{a k_b}(1 - e^{-k_b t}). \qquad (11)$$

Assuming $k_b = 0.042\ \mathrm{h}^{-1}$, Table 4 summarizes the solution (k_0) to Eq. (12) for the various flow rates. If $k_b = 0$, then,

$$C = C_0 - (2k_0/a)t, \qquad (12)$$

where $C \to 0$, as $t \to \infty$ and $C = 0$ for $t \geqslant C_0 a/2k_0$.
Figure 3 shows the relationship between the zero-order reaction rate coefficients versus mass transfer coefficient. These results are consistent with the findings of Digiano and Zhang (2005) findings that the reaction coefficient increases with mass transfer but approaches an asymptote as mass transfer increases (Clark et al. 2010).

6.3 Mass transfer limited model

In the Mass transfer limited model it is assumed that the chlorine concentration (c_w) at the pipe wall is zero due to instantaneous reaction. The flux expression then becomes:
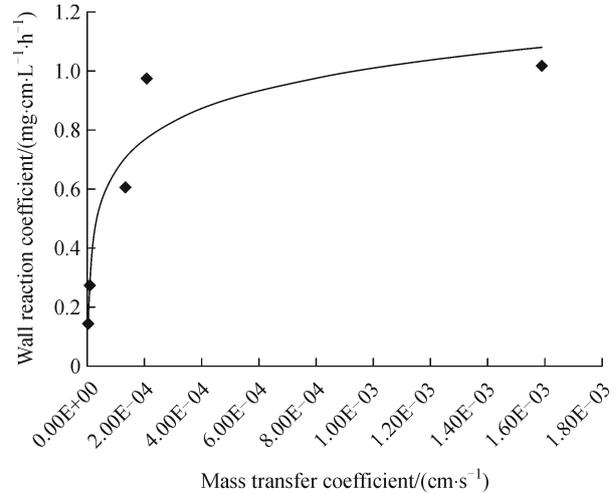
$$J_w = k_{mt}c. \qquad (13)$$

When substituted in the general transport equation (Eq. (7)), the mass transfer limited case becomes:

$$C = C_0 e^{-\left(k_b + \frac{k_{mt}}{a}\right)t}. \qquad (14)$$

However, this model tends to consistently underestimate the actual chlorine demand at low velocities (low mass transfer). It over estimates chlorine demand for higher velocities (high mass transfer) in the unlined metallic pipe.

7 Regulatory implications

It should be emphasized that the pipe material that makes-up the test loops may not be typical of the material that might be used in an operating drinking water distribution system. For example, the wall demand for the unlined metallic loops at the maximum flow rate was on order of 10/day in this study. However the basic principles developed from this study should be generally useful for all drinking water utilities. It is clear from the experiments reported in this paper that there are differences in the wall demand characteristics between unlined metallic pipe and PVC; the differences have major regulatory implications.

8 Conclusions

It has been hypothesized that internal corrosion of metal pipes can result in the deterioration of water quality; including the loss of chlorine residuals. Pipe wall demand for free chlorine has been attributed to a number of factors such as pipe age and material, pipe diameter, temperature, and pipe roughness. Recent studies have demonstrated that

in older unlined metal pipes, chlorine wall demand increases with flow rate. Digiano and Zhang (2005) observed, in their experiments, that cast-iron pipes exhibited zero-order pipe wall reactions, which increased with velocity due to mass transfer considerations. They found that steel pipes exhibited first-order reactions. Al-Jasser (2007) demonstrated that service age in both steel and cast iron pipes can increase wall demand.

To our knowledge this is the only reported set experiments in which "wall-demand" has been determined in parallel for two different types of pipe material simultaneously. The experimental results reported in this paper, demonstrate that surface or pipe wall demand varies significantly between different types of pipe material and, therefore, the type of pipe material can influence the transport and maintenance of free chlorine in drinking water distribution system networks. These characteristics can have regulatory implications; for example, chlorine residuals dissipate rapidly in unlined ductile-iron pipe but are maintained in PVC pipe. It is clear that pipe surface material has a major effect on wall demand. The authors believe that this is a phenomenon that should be studied more thoroughly.

It is also clear, from the experimental results, that the various models proposed for predicting chlorine transport have limitations. For example, based on the results presented in this paper, the wall demand parameter which is a fundamental parameter in EPANET and other water quality models is essentially a fitting parameter and is not necessarily constant under all flow conditions. This effect was also discussed in the paper by Clark and Haught (2005). The Zero-reaction model, frequently mentioned in the literature, has a similar problem. The zero-reaction coefficient also varies with flow, however, but as pointed out by Digiano and Zhang (2005), the coefficient varies in a more consistent and predictable manner. The mass transfer limited model has similar limitations. We believe this lack of fidelity becomes more serious as wall demand increases.

Despite some of the problems associated with the various types of models evaluated as part of this study, the authors believe that distribution system modeling is an important activity that should be maintained and expanded by water utilities. However, the authors also believe one of the lessons to be learned from this study is that no model is perfect and that it is critical for utilities to maintain a carefully constructed calibration program when distribution system models are used. It is clear that results from distribution system models can provide misleading information and they therefore should be applied with care and understanding.

Notations

a = pipe radius in cm

C_0 = initial chlorine concentration in mg/L

C = bulk flow chlorine concentration in mg/L

C_t = the concentration of chlorine in mg/L at time t

C_w = chlorine concentration at the pipe wall in mg/L

D_{ab} = molecular diffusion constant in cm^2/s

dc/dt = rate of change of concentration with respect to t

e = exponential

gpm = gallons per minute

J_w = radial flux in mg·cm/(L·s)

J_b = axial flux in mg·cm/(L·s)

K = the total first-order decay rate for chlorine in s^{-1}

k_0 = the zero-order reaction coefficient in mg·cm/(L·s)

k_r = the wall reaction constant (s^{-1})

k_w = first order coefficient for the pipe-wall reaction coefficient in cm/s

k_b = bulk decay coefficient in s^{-1}

k_{mt} = coefficient of mass transfer to the pipe wall in cm/s

ln = the natural log

t = time in second

u = advective velocity in the x direction in cm/s

x = the dimension along the pipe in cm

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