The decay of chlorine associated with the pipe wall in water distribution systems

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Abstract

Free chlorine decay rates in water distribution systems for bulk and wall demands should be modelled separately as they have different functional dependencies. Few good quality determinations of in situ wall demand have been made due to the difficulty of monitoring live systems and due to their complexity. Wall demands have been calculated from field measurements at 11 locations in a distribution system fed from a single source. A methodology for the laboratory determination has been evolved and shown to give results that are similar to the in situ results. Pipe materials were classified as either having high reactivity (unlined iron mains) or low reactivity (PVC, MDPE and cement-lined ductile iron). The results indicate that wall decay rates for the former are limited by chlorine transport and for the latter by pipe material characteristics. The wall decay rate is inversely related to initial chlorine concentration for low reactivity pipes. In general, water velocity increases wall decay rates though the statistical confidence is low for low reactivity pipes. A moderate biofilm coating did not influence the wall decay rate for low reactivity pipes. © 2002 Elsevier Science Ltd. All rights reserved.

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1. Introduction

Despite the effective use of chlorine over many decades as a disinfectant for potable water supplies, there are still regulatory failures in the quality of drinking water, which cause concern to the water utilities. An understanding of how chlorine concentrations will decay with time, due to reactions with trace compounds contained in the bulk water, material accumulated at the pipe wall or with the pipe material itself is, therefore, central to managing water quality. It is widely accepted that chlorine in distribution systems will decay, either due to reactions with compounds contained within the bulk water (bulk decay) or due to reactions at the pipe wall (wall decay). The simplest model to quantify chlorine decay in distribution systems uses a first-order decay equation with respect to chlorine:

$$dC/dt = -kc$$

or

$$C_t = C_0 \exp(-kt).$$

When considering in situ chlorine decay, the reactions associated with the bulk liquid are frequently separated from those associated with the pipe wall. Thus, AWWARF [1] defined the overall decay constant as the sum of a bulk constant and a wall constant:

$$k = k_b + k_w,$$

where k is the first-order decay constant (h^{-1}), k_b the bulk first-order chlorine decay constant (h^{-1}) and k_w the wall first-order chlorine decay constant (h^{-1}).

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This model has the advantages of simplicity and the decay constants, being in the same units, allow easy comparison. Measurements of the decay of chlorine associated with the pipe wall have been performed both in the laboratory and under field conditions. Powell et al. [2] reported the factors which control wall chlorine decay through field surveys of the in situ chlorine decay of a number of pipes in distribution.

Investigation into the in situ chlorine decay is still limited by the practical considerations such as estimation of travel times, identification of suitable sites and limits on the time to conduct such a survey. Most studies, therefore, consider only a small number of pipe lengths and focus on a small number of related parameters. Whilst in situ decay studies have advantages in being directly applicable to distribution, potential errors associated with the measurement may be large.

The decay rate is effectively calculated by observing the difference in chlorine concentration between two points and estimating the time of travel of the water between those points. Errors are, therefore, associated with the following [3]:

- Chlorine measurement: ± 0.01 mg/l by double Hach procedure.
- Travel time measurement: $\pm 15\%$ for flow meters; $\pm 5\%$ for tracer studies.
- Bulk measurement: ± 0.01 l/h.

A further error may be associated with iron pipes, which experience corrosion, reducing their effective internal diameter. This was estimated as -30% by Powell [3].

The pipe wall decay of chlorine has been measured in carefully controlled laboratory experiments, which aim at minimising the potential errors. Times corresponding to travel times can be carefully chosen, sample waters with little bulk decay can be used, the state of the internal surface of the test section of pipe can be observed and chlorine concentrations are easier to measure in laboratory conditions. The drawbacks of a laboratory study are that results may be less applicable and are often performed with new sections of pipe and under poor hydraulic regimes. The impact of parameters which influence the in situ decay is well documented and have been shown to have significant effects on the chlorine concentration in networks. The majority of research has been carried out using the first-order decay rate equation and the factors which have been shown to influence the wall decay are:

- pipe material and diameter [4–6];
- initial chlorine concentration [1];
- corrosion [1,7]; and
- biofilm [7,8].

The aim of this study was to consider wall decay as measured both in situ and under laboratory conditions

to develop a greater understanding of the factors which influence wall decay and to enable the results of laboratory studies to be more directly applied to distribution systems. The study aimed at examining a wider range of pipe types with more repeat wall decay determinations for in situ conditions, compared with work reported in the literature [6,9,10].

2. Materials and methods

2.1. Measurement of in situ wall decay

The in situ wall decay rate between any two points A and B was measured [3] as follows:

1. The bulk decay rate constant was measured at point A.
2. The average upstream or initial chlorine concentration, C_0, was measured for a slug of water at point A over ≈ 20 min.
3. The travel time, t, was calculated from point A to point B for that slug of water. This was obtained either from flow meter records or from tracer studies.
4. The downstream or final chlorine concentration, C, was measured as the slug of water arrived at point B. An average over ≈ 20 min was taken to ensure that the correct slug of water was measured. If a tracer study was used to calculate the travel time, then the period immediately prior to the trace being observed was used as the final chlorine concentration.
5. The overall and wall first-order decay rate constants were then calculated from

$$k = 1/t \times \ln C/C_0,$$
$$k_w = k - k_b.$$

Sites suitable for measurement of in situ wall decay were based on the following criteria:

- proximity to a chlorination point to ensure sufficient chlorine to measure;
- sufficient length to observe measurable decay;
- uniform pipe material and diameter to aid the interpretation of results; and
- safe and accessible sampling locations.

Flow meters could only be used where an accurate meter was directly connected to the surveyed pipe or to all connecting pipework, where all demands off the surveyed section were metered or were negligible and where the internal pipe diameter was accurately known. Tracer studies could only be used where a chlorine dosing unit or a flow meter where chlorine could be added was near to the surveyed section.

When measuring the in situ wall decay rate, the bulk demand was subtracted from the overall decay. To ensure a low bulk demand, the quarter strength Ringer's solution was used. Three bulk decay tests were carried out on Ringer's solution dosed with sodium hypochlorite and it showed a bulk decay rate (k_b) of $0.006 \, h^{-1}$ or less. This was deemed acceptable, being $<6\%$ of the measured average wall decay of $0.10 \, h^{-1}$ and less than the minimum recorded decay constant of $0.01 \, h^{-1}$.

All the in situ tests were conducted on the Melbourne fed, Leicester distribution system (Fig. 1) which supplies a total of 1.1 million customers using a lowland river as its source water. Between June 1996 and March 1999, 54 in situ decay rate measurements were undertaken from a total of 11 pipe sections (Table 1).

2.2. Measurement of laboratory-based wall decay

Laboratory-based wall demand tests were conducted on 450 mm length, 100 mm diameter sections of pipe

fitted between a PVC base plate and a perspex, conical-shaped lid. The instrument was fitted with an inlet valve entering towards the base, an outlet valve and an air release valve towards the top and a magnetic stirring facility (Fig. 2). The procedure for the test was as follows:

1. The instrument's base and lid were treated so that they were demand free by placing them in distilled water dosed to 10 mg/l free chlorine with concentrated hypochlorite for 24 h, before rinsing thoroughly with distilled water. This is the same procedure as used for the cleaning of equipment for the bulk decay rate.
2. A 10-l drum of quarter strength Ringer's solution was dosed to the desired chlorine concentration with concentrated hypochlorite solution and left to stand for 10 min to ensure homogeneity.
3. The demand instrument was filled by gravity through the inlet valve. As the instrument filled, the stirrer
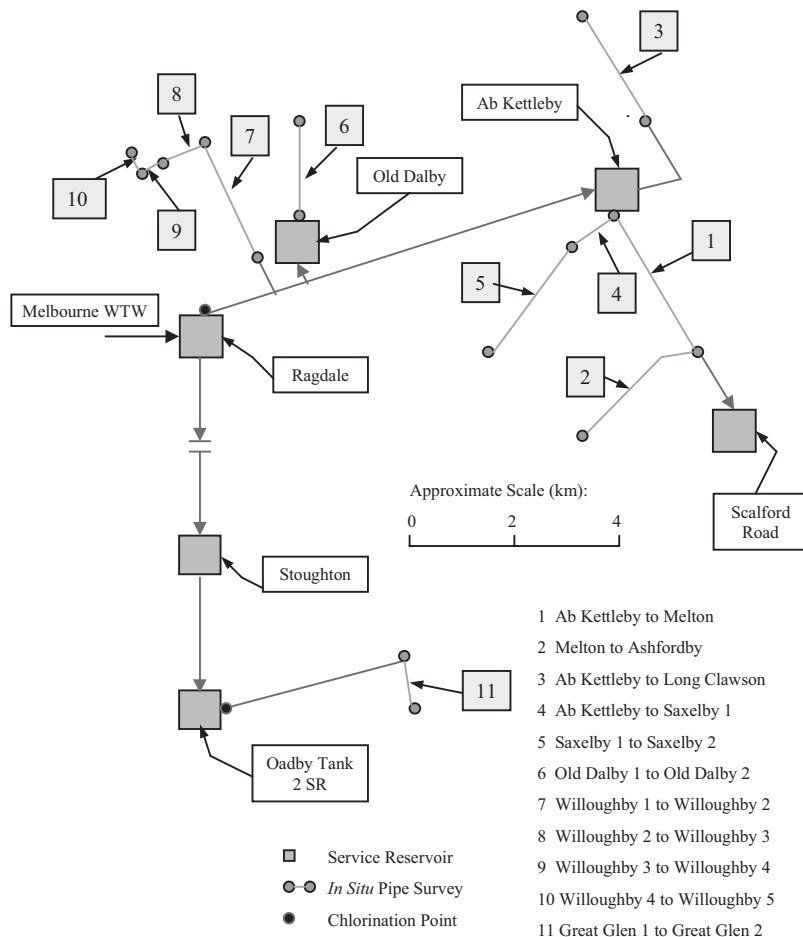

Fig. 1. Schematic of the in situ wall decay test locations (all fed from Ragdale service reservoir).

Table 1
Summary of the in situ wall decay tests

| Location	Pipe characteristics				
	No. of tests	Diameter (mm)	Length (m)	Material	Year laid
Ab Kettlby/Melton	7	298	3505	DICL	Relined 1991
Melton/Ashfordby	7	180	2610	MDPE	1985
Ab Kettlby/Long Clawson	6	150	2300	PVC	1979
Ab Kettlby/Saxelby 1	5	200	1030	SI	1959
Saxelby 1/Saxelby 2	5	200	2284	SI	1959
Old Dalby Res./Old Dalby	3	150	2022	CI	Pre-1960
Willoughby 1/Willoughby 2	6	150	2165	PVC	1973
Willoughby 2/Willoughby 3	7	100	925	CI	1960
Willoughby 3/Willoughby 4	4	100	515	CI	1960
Willoughby 4/Willoughby 5	2	100	305	CI	1960
Great Glen 1/Great Glen 2	2	200	865	PVC	1991
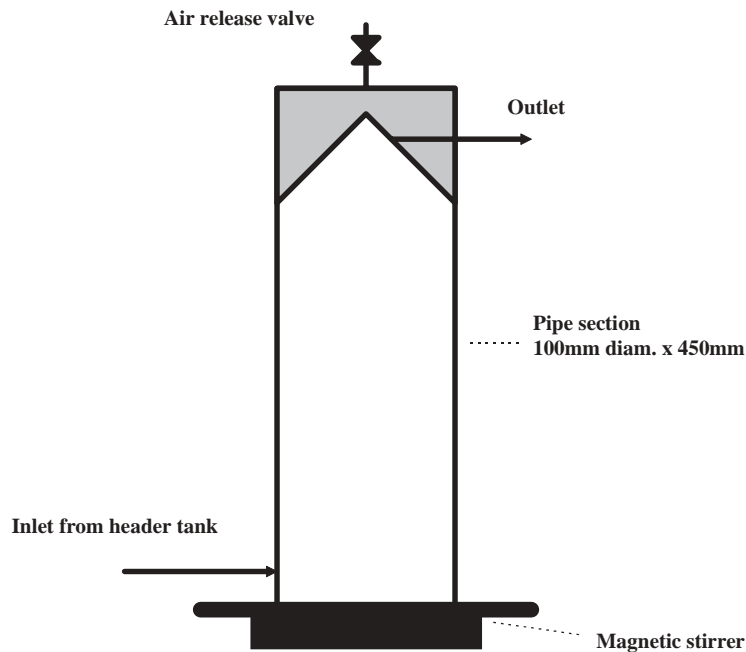
Fig. 2. Schematic diagram of the laboratory-based wall decay monitor.

was turned on to ensure continuous renewal of water at the pipe surface.

4. As the instrument became full, a certain volume (20 ml) of water was run to waste before the first sample was taken from the outlet valve using the double Hach procedure [11]. All water released was replaced from the original drum by gravity.

5. The air valve was opened and a small volume of water released (<5 ml) to allow any trapped air to escape.

6. Further samples were taken in the same manner at intervals throughout the period of the test. Ideally, samples were taken at times to ensure a regular proportional drop in chlorine.

78% of the tests were carried out on pipe sections, which had not been in the system and were therefore considered 'new'. They were treated, prior to testing, by submerging in distilled water dosed to 10 mg/l free chlorine with hypochlorite solution for 48 h and rinsed with distilled water. A further 20% of the tests were carried out on 'new' pipe sections, which had been placed within the system with water passing through for

a period of 3 weeks in order to produce a biofilm. The pipe was placed into the instrument while the surface was still wet, ensuring that the state of any biofilm was not disturbed. The remainder of the tests (2%) were carried out on pipe sections which had been within the system for a number of years. These were cut out and the biofilm, which had dried during transportation, was rinsed with distilled water to moisten it before the section was placed in the demand instrument. The authors accept that this technique disturbs the inside of the pipe to some degree. However, measurement on site was considered impractical.

Six pairs of laboratory-based wall decay tests were carried out on the same section of pipe under the same conditions of initial chlorine dose and temperature to determine the reproducibility of the tests. Results showed that the standard deviation of the errors was $\pm 0.013\,h^{-1}$ which, when analysed assuming a Student's t distribution, gave a standard error of $\pm 0.037\,h^{-1}$ and 95% confidence limits of $\pm 0.016\,h^{-1}$.

A number of differences exist between the laboratory model and the field tests, which affect the associated errors. In the laboratory studies:

- In the majority (98%) of cases, new pipes were used, eliminating the impact of the age and condition of the pipes.
- The hydraulic regime was only an approximation of system conditions. The magnetic stirrer used to ensure water movement was temperamental. However, tests concluded that its use had no significant impact on wall decay.
- Water samples were taken from the top of the monitor through a sample line away from the pipe wall. Samples were, therefore, not taken at the wall but from the bulk liquid.
- Initial chlorine concentrations in the laboratory experiment were greater than observed in distribution systems. Higher concentrations ensured that any small errors from the pocket colorimeters were minimized.
- Water temperature of the tests averaged 19°C, warmer than under most field conditions.
- Laboratory tests were conducted over a longer time period than field tests in order to ensure that the majority of chlorine in the water had decayed.

2.3. Tracer studies

Good hydraulic information is vital in considering chlorine decay within a system in order to gather realistic travel time information. Flow meters within a system are often limited as they are expensive to install and their accuracy is often difficult to assess. Pressure monitoring provides an alternative. However, this is only accurate where pressure gradients are high and pipework diameters and friction factors are known. Tracer studies offer the flexibility to determine travel times to virtually any point in the system.

Chlorine tracing was carried out using the following methodology:

1. *Either* chlorine was injected into the network to produce a spike of chlorinated water. The spike was in the range of 0.5–1.0 mg/l free chlorine for a duration of 15 min. This was achieved either by increasing the target on a nearby chlorination equipment or by connecting an emergency-dosing equipment to the mains via a universal cock where a flow meter existed.

   Or the chlorine-dosing equipment was turned off for a period of 30 min producing a significant dip in free chlorine concentration. If the usual chlorine concentrations would allow, this method avoided using any high concentration of chlorine in the system.
2. Hydrant sampling was implemented downstream of the chlorine injection sites, either with a continuous monitor (EIT Model 5150), or with the personnel continually taking spot samples. The chlorine spikes, or dips, were, thus, followed down a length of main into the system. Hydrant flows were set to 6 l/min or less in order to minimise disruption to the natural flow in the main.
3. The travel time between two points was calculated as the period between the rising limb of the spike and falling limb of the dip. This was chosen so that the equipment could, immediately, be moved downstream if circumstances necessitated.

3. Results and discussion

The in situ wall decay constants varied from 0.00 to $1.64\,h^{-1}$. 70% of the constants were $<0.4\,h^{-1}$ and an examination of Fig. 3 shows that most of the sites surveyed had low decay constants. However, five sites were associated with some higher ($>0.4\,h^{-1}$) values and two sites; Willoughby 3/Willoughby 4 and Old Dalby Reservoir/Old Dalby; gave decay constants which were all $>0.4\,h^{-1}$. Both of these were cast iron (CI) mains, the former with a nominal diameter of 100 mm, the latter with one of 150 mm.

The laboratory experiments were conducted on new polyvinyl chloride (PVC), cement lined cast iron (CICL) and medium density polyethylene (MDPE) pipe sections. The wall decay constants ranged from $0.01\,h^{-1}$ to 0.78 l/h (Fig. 4). These results showed a slightly greater range of decay rates than the in situ tests on the same materials, which ranged from 0.00 to $0.26\,h^{-1}$.

The average in situ decay of CI and spun iron (SI) pipes surveyed was significantly greater than the

cement-lined ductile iron (DICL), PVC and MDPE pipework (Fig. 5). Chlorine is a strong oxidising agent, which reacts readily with iron. However, the variability in the cast iron decay constants was large, varying between 0.03 and $1.64 \, h^{-1}$. This variation will, in part, be related to the condition of the pipe itself. The average wall decay of the DICL ($0.13 \, h^{-1}$) main was slightly higher than those for the PVC ($0.09 \, h^{-1}$) or the MDPE

($0.05 \, h^{-1}$) mains, though still significantly lower than unlined iron pipes. Therefore, the pipe reactivity, as judged by the wall decay constants, was found to be CI > SI > DICL > PVC > MDPE. It is clear that the cement provides a layer of protection for the iron against attack by chlorine. Previous in situ studies have also shown that unlined cast iron pipes have decay rates between 4 and 100 times greater than lined or plastic pipes [1].

For the laboratory studies, the pipes were separated into relatively reactive pipes (CI and SI) and relatively unreactive pipes (MDPE, PVC and CICL). Fig. 6 shows that laboratory experiments produced similar results to the field tests, suggesting a good relationship between the two methods of measuring wall decay. The average wall decay as measured in laboratory and field tests for the different materials, respectively, were 0.12 and
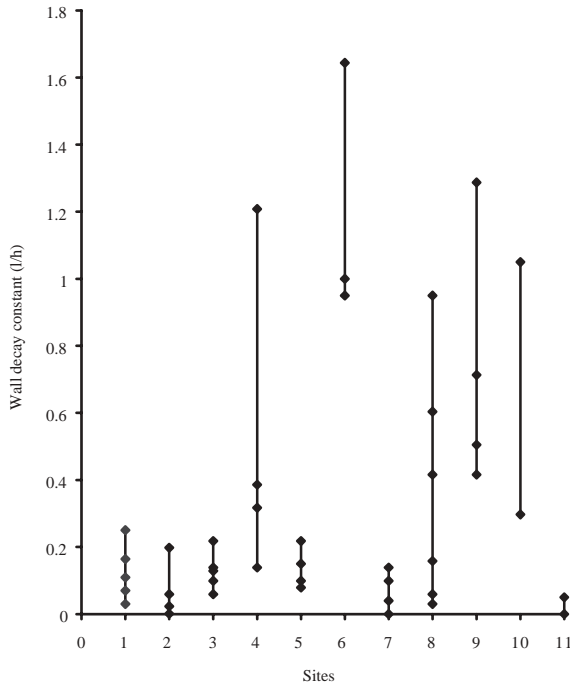

Fig. 3. In situ wall decay constants at the various sites (as numbered in Fig. 1).
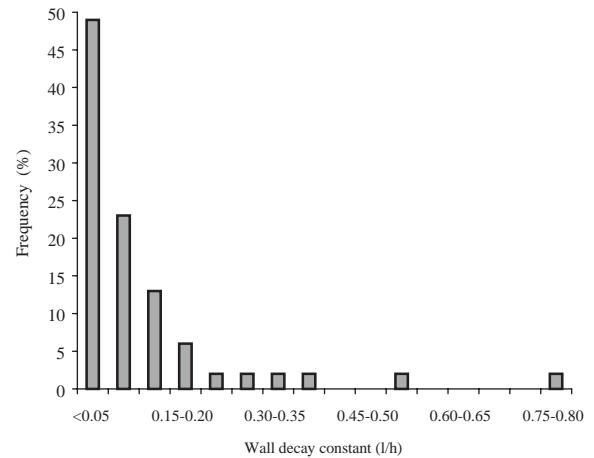

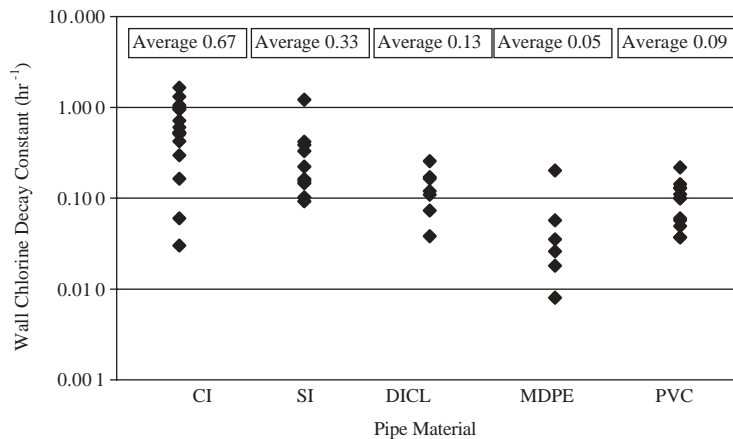Fig. 4. Variation in the laboratory wall decay constant.

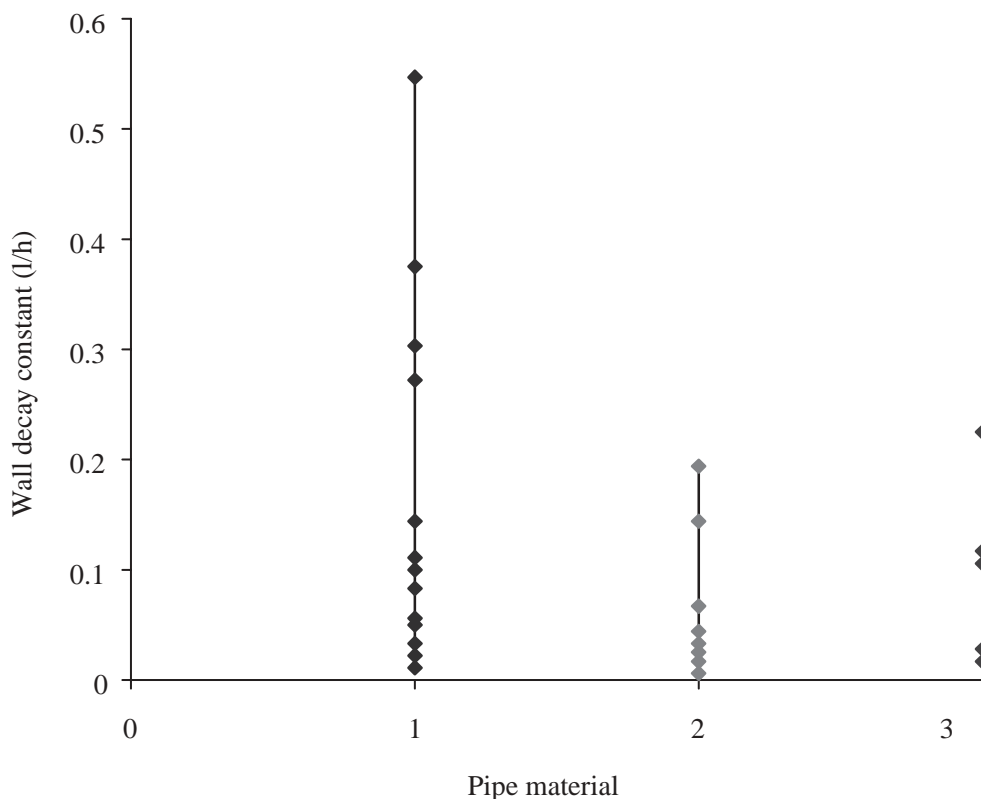Fig. 5. Effect of pipe material on wall decay constant k_w.

Fig. 6. The impact of pipe material on the laboratory-measured wall decay constant (1—CICL; 2—MDPE; 3—PVC).

$0.13\,h^{-1}$ (CICL and DICL), 0.09 and $0.09\,h^{-1}$ (PVC) and 0.05 and $0.05\,h^{-1}$ (MDPE). Laboratory studies have shown that new plastic pipes have almost no chlorine demand, other than a small initial demand on first contact with chlorine, which would be satisfied very quickly in the system [5].

The bulk chlorine decay was observed to have an inverse relationship with the initial chlorine concentration. A similar inverse relationship with wall decay on one pipe stretch dosed to between 0.3 and $5.0\,mg/l$ has been reported by AWWARF [1] but stabilisation of the wall decay, following a change in initial chlorine concentration, took ≈ 1 month. In the current study, a strong relationship between initial chlorine concentration and wall decay for all PVC pipes surveyed in situ was found. The regression equation shown in Fig. 7 is $y = 0.26 - 0.73x$ and analyses showed that the relationship was significant at the 99% level. This suggests that the wall decay constant would be zero at initial chlorine concentrations $> 0.36\,mg/l$. The logical proposition would be that wall decay would tend to, but not reach, zero. However, neither exponential nor power functions adequately fitted this data. The linear function proposed in Fig. 7 should, therefore, only be considered representative between initial chlorine concentrations of 0.15 and

$0.3\,mg/l$. Similar relationships were not evident for MDPE, DICL or iron pipes. It is possible that the small range of initial chlorine concentrations studied was responsible. Alternatively, there might have been insufficient time for stabilisation of the wall decay constant to occur, as indicated by the work reported previously [1]. If either of the above explanations is true, however, it does pose the question why was the relationship evident for PVC pipes?

In the laboratory experiments, with samples being dosed to initial chlorine concentrations of between 0.16 and $1.95\,mg/l$, relationships between initial chlorine concentration and wall decay constants were also observed (Fig. 8). A linear function was not found to fit the data adequately. A power function provided the best fit when applied to each material type (Table 2). The relationships were all significant at the 99% level. The inverse relationship suggests that there is a limiting rate at which the chlorine can be taken up by the pipe wall, hence higher chlorine concentrations yield lower k_w values. Unlined iron pipes, with a highly reactive surface, would not be expected to be limited in the same way, hence it is not unreasonable that no such relationship was found with the in situ measurements under this type of pipe. No such comparison was
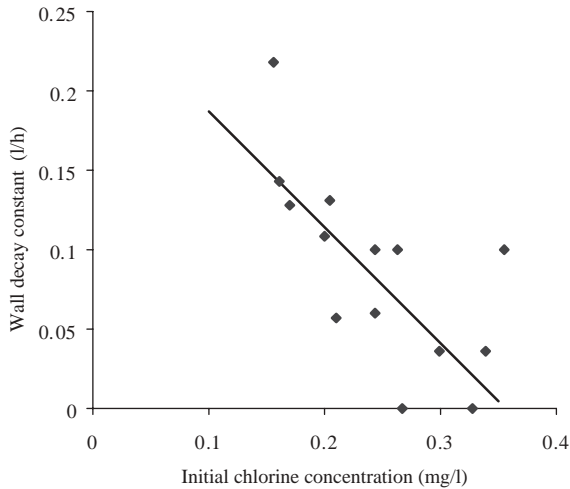
Fig. 7. Relationship between the initial chlorine concentration and the in situ wall decay constant for PVC pipes.
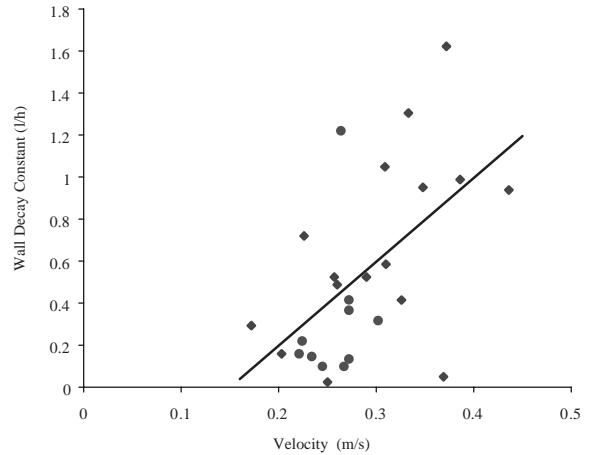

Fig. 9. Relationship between velocity and the in situ wall decay constants for SI (●) and CI (◆) pipes.
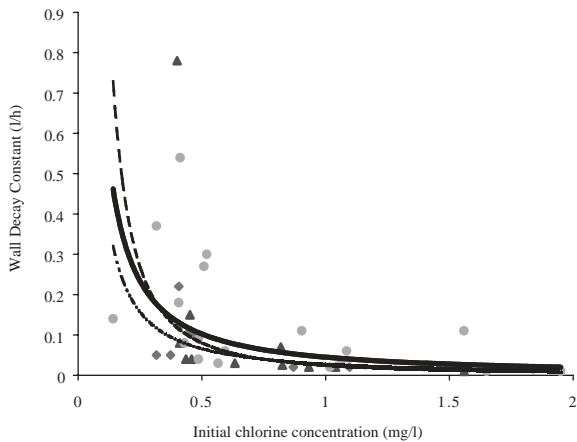

Fig. 8. Relationships for initial chlorine concentration and the laboratory-measured wall decay constant for CICL (● ━━━), MDPE (▲ ━ ━ ━) and PVC (◆ ━━ · ━━).

Table 2
Regression equations for the relationships between initial chlorine concentrations and the wall decay constant for different pipe materials

Pipe material	Regression equation	R^2
CICL	$y = 0.05x^{-1.17}$	0.48
PVC	$y = 0.02x^{-1.55}$	0.67
MDPE	$y = 0.02x^{-1.75}$	0.52

conducted under laboratory conditions as no pipes were available for testing.

The hypothesis that the rate at which chlorine can be transferred to the pipe wall was limiting decay, due to either turbulence or diffusion, was rejected because, under high chlorine concentrations, a greater concentration difference would exist between the pipe center and the pipe wall where the majority of decay was occurring. This would increase the driving force controlling the rate at which chlorine would diffuse to the pipe wall. Hence, it would be expected that greater chlorine concentrations would increase, not decrease, transport to the pipe wall and in turn decay. Turbulence would be the same under any chlorine concentration.

Flow velocity can affect turbulence, diffusion and the boundary layer thickness, all of which influence the rate at which chlorine is transferred to the pipe wall. A positive relationship would be expected where transfer of chlorine to the pipe wall is limiting as velocity increases the rate at which chlorine would be transferred. Fig. 9 shows that there was a linear relationship between flow velocity and the in situ decay constants for unlined iron pipes. This relationship was significant at the 99% level. This linear equation suggests that if velocity were reduced below 0.15 m/s in unlined iron pipes, wall decay would be eliminated. This is unrealistic, as some transfer of chlorine would still occur resulting in positive wall decay. A linear response is, therefore, perhaps fortuitous. However, neither an exponential nor a power function was found to represent the data adequately. This relationship should, therefore, only be considered representative of velocities between 0.2 and 0.45 m/s.

A similar positive relationship was found for PVC, MDPE and DICL pipes. However, the fit was poor ($R^2 = 0.26$), as might have been be expected with unreactive pipes where the pipe reactivity and not transfer of chlorine would be rate limiting.

The flow in all the pipes surveyed was in the transitional region of the Moody diagram, with the

Reynolds number ranging by an order of magnitude from 1.7×10^4 to 1.7×10^5.

The relationships determined between the Reynolds numbers and the decay constants indicated that turbulence had a similar effect on wall decay as on velocity. Generally, the relationships were poor, particularly for SI, PVC, MDPE and DICL pipes.

Pipe diameter is inversely proportional to the surface to volume ratio. The higher the surface to volume ratio, the larger the surface area across which mass transfer can occur and the greater the number of reactive sites available for each unit volume of water passing down the pipe. Previous research has observed decay rates to decrease with pipe diameter [5,6]. Measuring the impact of diameter on in situ chlorine decay was difficult, as, ideally, pipe stretches with differing diameters but otherwise identical properties were needed. As can be seen from Table 1, these criteria did not exist in the survey area. No relationship was evident for any pipe type, although only a small range of pipe diameters was considered.

Temperature will affect a range of parameters, which can influence chlorine decay. The data from the surveys suggested that a higher wall decay did occur at higher temperatures, but the relationships were such that no conclusions could be drawn. The TOC data showed a considerable amount of scatter and, again, no conclusions could be drawn about its effect on the wall decay constants.

A biofilm, which develops on the internal surface of a pipe may create a chlorine demand itself or it may protect a surface from chlorine penetration, thus reducing decay.

The only previous research on the chlorine demand of a biofilm was conducted by Lu et al. [8] and Kiene et al. [7], who developed a biofilm on 4 mm polystyrene beads and measured the resulting demand. The biofilm was shown to exert a significant but small chlorine demand of ≈ 0.11–$0.18 \, h^{-1}$ for colonized beads and $0.10 \, h^{-1}$ for uncolonized beads. No measure of the biofilm quantity on the beads was given. A limitation of this work is that it considers the biofilm grown on polystyrene beads and not on the actual sections of pipe and ignores the interaction between the material and biofilm. The laboratory monitor was, therefore, used to measure the chlorine decay due to a biofilm in contact with a pipe surface.

The same pipe sections used to assess the effect of pipe material on the the chlorine demand (PVC, MDPE and CICL) were placed within parts of the distribution system as vertical pipe reactors [12] and a biofilm were allowed to develop. The amount of biofilm on the pipe sections measured for chlorine decay ranged from 51 to 670 pg ATP/cm^2. This moderate quantity of biofilm did not significantly influence the decay of chlorine at the pipe wall. 12 samples were tested when biofilm was present on the pipe surface. Only one of the samples showed a decay constant, which was outside the range of values obtained when no biofilm was present. These results, perhaps, go some way to explaining why TOC had no influence on wall decay. A larger amount of biofilm, as has been reported by some authors [13,14], may have a more significant effect on chlorine decay.

4. Conclusions

- An acceptable method for measuring the in situ chlorine decay in distribution was developed. A laboratory experiment to measure the wall decay gave results representative of the distribution system.
- The pipe material has a strong influence on the wall decay in distribution. Pipes can generally be classified into reactive pipes (unlined iron) and unreactive pipes (PVC, MDPE, cement-lined iron).
- Results suggest that the wall decay of unreactive pipes is limited by the reactivity of the pipe while reactive pipes are limited by the transfer of chlorine to the pipe wall.
- An inverse relationship was found over a small observed C_0 range (0.15–0.3 mg/l) for in situ PVC pipes and for unreactive pipes in laboratory tests over a wider range of C_0 (0.16–1.95 mg/l).
- A positive relationship existed between flow velocity and wall decay for all pipe sections surveyed. The relationship between Reynolds number and wall decay is less significant
- Relationships were not observed between wall decay and the parameters of TOC, pipe diameter and temperature. There is a suggestion, however, that increased temperatures may make the decay more variable.
- A moderate amount of biofilm (51–670 pg ATP/cm^2) on the wall of unreactive pipes did not influence the wall decay rate.

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